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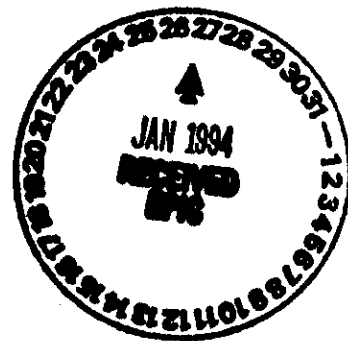
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## 100 Area Soil Washing Bench-Scale Tests

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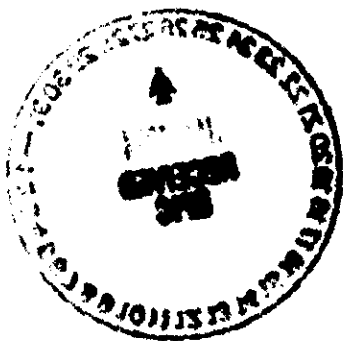


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## ACRONYMS AND ABBREVIATIONS

ASTM	American Society of Testing Materials
BSE	Back-Scattered Electrons
CD	Citrate-Dithionite
CEC	Cation Exchange Capacity
CERCLA	Comprehensive Environmental Response, Compensation, and Liability Act
DCB	Dithionite-Citrate-Bicarbonate
DOE-RL	Department of Energy, Richland Operations Office
DOT	Department of Transportation
EPA	Environmental Protection Agency
FS	Feasibility Study
HAH	Hydroxyl Amine Hydrochloride
HP	Horse Power
ICP-MS	Inductively-Coupled Plasma Mass Spectrometry
JCPDS	Joint Committee on Powder Diffraction Standards
MDA	Minimum Detectable Activity
NPL	National Priorities List
NTU	Nephelometric Turbidity Units
PNL	Pacific Northwest Laboratory
RI	Remedial Investigations
RPM	Revolutions Per Minute
SE	Secondary Electrons
SEM-EDS	Scanning Electron Microscopy-Energy Dispersive Spectrometry
TCLP	Toxicity Characteristic Leaching Procedure
TOC	Total Organic Carbon
TPL	Target Performance Level
WHC	Westinghouse Hanford Company
XRD	X-Ray Diffraction
XRF	X-Ray Fluorescence

## EXECUTIVE SUMMARY

The Pacific Northwest Laboratory conducted a bench-scale feasibility study on two 100 Area contaminated soils. The objective of this study was to evaluate the use of physical separation systems and chemical extraction methods as a means of separating chemically and radioactivity-contaminated soil fractions from uncontaminated soil fractions. The feasibility study was conducted on soil samples from two trenches, 116-C-1 and 116-D-1B. Two samples of soil were obtained from trench 116-C-1: one from the middle of the trench (Batch I) and one near the inlet (Batch II). A single sample (Batch III) was obtained from trench 116-D-1B.

Particle size distribution data indicated that <2-mm-sized fractions constituted approximately 10%, 2.8%, and 53.1% of the total mass in Batches I, II, and III, respectively.

The pH of the soils ranged from 6.5 to 7.66, and total organic carbon contents were relatively low (0.06 to 0.164 %). The cation exchange capacities of all three soils were typical of Hanford soils, ranging within the narrow limits of 8 to 8.9 meq/100 g. In all cases, the dominant exchangeable cation was Ca. Trace element analyses of these soils (<2 mm-sized fractions) showed no anomalies. Total chromium concentrations in these soils were relatively low (56 to 236 mg/kg). The Toxicity Characteristic Leaching Procedure extraction of these soils indicated that all eight inorganic constituents (silver, arsenic, barium, cadmium, chromium, mercury, and selenium) were well below the regulatory levels.

The radionuclide data indicated that the soil-washing tests should be focused on Batch II from trench 116-C-1 and Batch III from trench 116-D-1B. All soil-washing tests on Batch II material were conducted on the coarse fraction (>2 mm) because it was the major fraction (97.2%) of this soil. In contrast, the radionuclide data for Batch III showed that it would be appropriate to conduct additional soil-washing tests on the 2- to 0.25-mm-sized fraction (42.3% of the total soil mass in Batch III). In these soil fractions the activities of only  $^{60}\text{Co}$ ,  $^{137}\text{Cs}$ , and  $^{152}\text{Eu}$  exceeded the target performance levels (TPL). Therefore, the effectiveness of subsequent soil-washing tests for Batch III were evaluated on the basis of activity attenuation of these radionuclides in the 2- to 0.25-mm-sized particle fractions.



Two types of tests (physical and chemical) were conducted to reduce the activities of  $^{60}\text{Co}$ ,  $^{137}\text{Cs}$ , and  $^{152}\text{Eu}$  in the soil fractions of Batches II and III. The physical tests consisted of attrition scrubbing (2- to 0.25-mm-sized fraction of Batch III) and autogenous grinding (> 2 mm-sized fraction of Batch II). Chemical extractions were also conducted on both samples.

Preliminary attrition-scrubbing tests indicated that for Batch II (2 to 0.25-mm fraction), the optimum pulp density for effective scrubbing was approximately 83%. Based on these data, a number of attrition scrubbing tests were conducted to establish the relationship between energy input, reduction in radionuclide activity, and the amount of fines (< 0.25-mm material) generated. The results indicated optimum performance was obtained at an average energy input of 1.43 HP-min/lb (residence time 30 min). The average reductions in  $^{60}\text{Co}$ ,  $^{137}\text{Cs}$ , and  $^{152}\text{Eu}$  activities were > 80%, 28%, and 61%, respectively. Scrubbing at this intensity generated about 9% by weight fines. Doubling the energy input did not result in any noticeable increase in radionuclide removal efficiencies. The results of another scrubbing experiment with an electrolyte solution (0.5M ammonium citrate with citric acid) showed > 79%, 39%, and 83% reduction in  $^{60}\text{Co}$ ,  $^{137}\text{Cs}$ , and  $^{152}\text{Eu}$  activities. Such enhanced radionuclide removal by electrolyte addition appeared to be a result of the synergistic combination of scrubbing action, the improved dissolution of radionuclide-bearing surface coatings, and the reduced readsorption of solubilized contaminants onto freshly exposed surfaces of soil grains. Optical and electron microscopic observations of soil particles before attrition scrubbing indicated the presence of iron oxide and aluminosilicate coatings on the particle surfaces. Observations of the scrubbed particle surfaces showed significant removal of these coatings, indicating that major fractions of  $^{60}\text{Co}$  and  $^{152}\text{Eu}$  and a minor fraction of  $^{137}\text{Cs}$  were associated with the surface coatings.

Two-stage attrition scrubbing experiments were also conducted on Batch III (2- to 0.25-mm-sized fraction). The results show that two-stage scrubbing with deionized water removed greater amounts of radionuclides than single-stage scrubbing with deionized water. Among all scrubbing experiments, the removal of radionuclides was highest when two-stage scrubbing was conducted with an electrolyte present in both stages. During this two-stage process, > 79%, 48%, and 94% of  $^{60}\text{Co}$ ,  $^{137}\text{Cs}$ , and  $^{152}\text{Eu}$  activity was removed, and about 14% by weight fines was generated.

The results of autogenous grinding experiments conducted on coarse fractions (> 2 mm-sized) from Batch II showed that 88% and 94% of  $^{60}\text{Co}$  and  $^{152}\text{Eu}$  activities, respectively, could be removed if the grinding was conducted with 17% electrolyte. The measured residual activities of these two radionuclides after grinding were below the TPL. In contrast, grinding resulted in only a 25% reduction in  $^{137}\text{Cs}$  activity. Approximately 5% by

weight fines was generated during this experiment. Additional dry and wet (with deionized water) grinding with 17% to 25% sand as a grinding medium resulted in radionuclide removal in amounts comparable to the grinding experiment with electrolytes but resulted in the formation of substantial amount of fines (13% to 19% by weight).

Chemical extraction experiments were also conducted on Batch III (2- to 0.25-mm-sized fraction), and Batch II (>2-mm-sized fraction). Preliminary extraction studies were conducted with widely used extract compositions. These extractants consisted of a mineral acid (HCl), two organic acids (acetic, citric) with and without reducing agents (sodium dithionite, hydroxylamine hydrochloride), and a neutrally-buffered reductive solution (sodium bicarbonate, dithionite, citrate). These extractions were conducted at ambient or extractant-specific temperatures (80 to 96°C). Even though some extractants removed major fractions (>90% and 92%) of  $^{60}\text{Co}$  and  $^{152}\text{Eu}$ , they were less effective in reducing  $^{137}\text{Cs}$  activities (30% to 40% removal). Therefore, based on the knowledge of contaminant-substrate association, two new extractants were formulated and tested on these soil fractions. These new extractants removed major fractions of all three radionuclides from Batch III, consistently removing an average of >90%, 85%, and >99%, of  $^{60}\text{Co}$ ,  $^{137}\text{Cs}$ , and  $^{152}\text{Eu}$ , respectively, resulting in residual activities that were well below the TPL.

Chemical extraction of the gravel fraction from Batch II with one of the new extractants was conducted under static conditions. Significant reductions in  $^{60}\text{Co}$  (82%), and  $^{152}\text{Eu}$  (92%) activities resulted in residual activities below the TPL were observed in these experiments. However, the reduction in  $^{137}\text{Cs}$  activity never exceeded 40%, and because of the inherently higher activity of  $^{137}\text{Cs}$  in the coarse material from Batch II, the overall residual activity was still elevated. Removal of  $^{137}\text{Cs}$  (57%) was increased in another autogenous grinding experiment with coarse material from Batch II in contact with one of the new extractants.

Preliminary flocculation tests were conducted on waste-water streams generated from wet-sieving, two-stage attrition scrubbing with an electrolyte, and chemical extraction (extractant II) of 116-D-1B (Batch III) soil. These tests showed that it was feasible to flocculate the suspended solids in these liquid wastes by using commercially available flocculents (CATFLOC-L, POL-E-Z 692). The activities of  $^{60}\text{Co}$ , and  $^{152}\text{Eu}$  in supernatants from treated or untreated waste solutions were at or below minimum detectable activities (MDA) of 1.7 and 4.3 pCi/ml respectively. The activity of  $^{137}\text{Cs}$  in supernatant from spent chemical extraction solutions were measured at 6 pCi/ml (MDA: 1.6 pCi/ml).

The data obtained from these feasibility tests indicate that even though soil-washing significantly reduced the activities of  $^{60}\text{Co}$  and  $^{152}\text{Eu}$  in Batch II, the activity of  $^{137}\text{Cs}$  could not be substantially reduced. Finally, the tests on Batch III show that the soil from trench

116-D-1B can be successfully soil washed by combining wet sieving with either two-stage attrition scrubbing in electrolyte or a single chemical extraction step. The anticipated mass reduction of contaminated material for this soil is approximately 84% to 87%.

Based on this study, the feasibility of soil-washing each of the three soils are assessed as follows:

- 116-C-1 (Batch I) Soil: The activities of radionuclides in bulk soil are all below TPL therefore, soil-washing appears unnecessary and no specific treatment plan has been provided.
- 116-C-1 (Batch II) Soil: The residual activity of  $^{137}\text{Cs}$  in all treated soils were above TPL. Because the soil-washing treatments tested in this study were not effective enough to attain the TPL for this radionuclide, no specific treatment plan has been provided.
- 116-D-1B (Batch III) Soil: Soil-washing tests were effective in reducing the radionuclide activities in bulk soil to below TPL. Therefore, a specific treatment plan has been recommended (See 11.0).

Finally, this study showed that choosing appropriate soil washing schemes may lead to successful remediation of soils in many of the 100 Area waste sites.

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## Contents

EXECUTIVE SUMMARY .....	iv
1.0 INTRODUCTION .....	1-1
2.0 SOIL-WASHING TECHNOLOGY .....	2-1
2.1 DESCRIPTION OF THE TECHNOLOGY .....	2-1
2.2 FEASIBILITY OF SOIL-WASHING RADIONUCLIDE- CONTAMINATED SOILS .....	2-2
2.3 TARGET PERFORMANCE LEVELS FOR RADIONUCLIDES ...	2-4
2.4 A SOIL-WASHING TEST SCHEME FOR 100 AREA SOILS .....	2-4
3.0 SOIL SAMPLES FROM 100 AREA .....	3-1
3.1 SITE DESCRIPTION .....	3-1
3.2 TEST SITES .....	3-1
3.3 SOIL SAMPLE COLLECTION .....	3-6
3.4 SOIL SAMPLE PREPARATION .....	3-6
4.0 CHARACTERIZATION OF SOILS .....	4-1
4.1 PHYSICAL CHARACTERIZATION .....	4-1
4.1.1 Moisture Content .....	4-1
4.1.2 Particle Size Distribution .....	4-1
4.1.3 Specific Gravity .....	4-1
4.2 CHEMICAL CHARACTERIZATION .....	4-2
4.2.1 Total Organic Carbon .....	4-2
4.2.2 Soil pH .....	4-2
4.2.3 Cation Exchange Capacity .....	4-2
4.2.4 Total Elemental Analyses .....	4-2
4.2.5 Radionuclide Analyses .....	4-3
4.2.6 Toxicity Characteristic Leaching Procedure (TCLP) .....	4-3
4.2.7 Sequential Extraction .....	4-3
4.3 MINERALOGICAL CHARACTERIZATION .....	4-4
4.4 RESULTS AND DISCUSSION .....	4-5
4.4.1 Physical Characterization .....	4-5
4.4.2 Chemical Characterization .....	4-5
4.4.3 Mineralogical Characterization .....	4-13
5.0 WET-SCREENING .....	5-1
5.1 OBJECTIVE .....	5-1
5.2 EQUIPMENT AND PROCEDURES .....	5-1
5.3 RESULTS AND DISCUSSION .....	5-3

94/3221.0010

## Contents (cont.)

6.0	ATTRITION SCRUBBING .....	6-1
6.1	OBJECTIVE .....	6-1
6.2	EQUIPMENT AND PROCEDURES .....	6-1
6.3	RESULTS AND DISCUSSION .....	6-3
7.0	AUTOGENOUS GRINDING .....	7-1
7.1	OBJECTIVE .....	7-1
7.2	EQUIPMENT AND PROCEDURES .....	7-1
7.3	RESULTS AND DISCUSSION .....	7-2
8.0	CHEMICAL EXTRACTION .....	8-1
8.1	OBJECTIVE .....	8-1
8.2	EQUIPMENT AND PROCEDURES .....	8-1
8.3	RESULTS AND DISCUSSION .....	8-2
9.0	COMBINATION TESTS .....	9-1
9.1	OBJECTIVE .....	9-1
9.2	DESCRIPTION OF TESTS .....	9-1
9.3	RESULTS AND DISCUSSION .....	9-2
10.0	WASTE WATER TRETMENT .....	10-1
10.1	OBJECTIVE .....	10-1
10.2	EQUIPMENT AND PROCEDURE .....	10-1
10.3	RESULTS AND DISCUSSION .....	10-2
11.0	DISCUSSION AND RECOMMENDATIONS .....	11-1
11.1	CONTAMINANT RADIONUCLIDES IN 100 AREA SOILS .....	11-1
11.2	SOIL-WASHING PARAMETERS .....	11-2
11.3	RESULTS OF THE FEASIBILITY TESTS .....	11-2
11.4	SOIL-WASHING PROCESS OPTIONS FOR 116-D-1B SOILS .....	11-4
11.4.1	Physical Soil-Washing Process Options .....	11-4
11.4.2	A Preliminary Soil-Washing Scheme for 116-D-1B Soil .....	11-7
11.5	PROPOSED PILOT-SCALE SOIL-WASHING TESTS ON 116-D-1B SOIL .....	11-9
11.6	PRELIMINARY COST ESTIMATES FOR SOIL WASHING OPTIONS .....	11-9
11.7	SUPPLEMENTARY DATA REQUIREMENTS .....	11-10
12.0	REFERENCES .....	12-1
	Appendix A - Cost Estimates .....	A-1

**Figures**

1-1	The Location of the Hanford Site. . . . .	1-2
2-1	A Scheme for Soil-Washing Feasibility Tests on 100 Area Soils . . . . .	2-5
3-1	Generalized Geologic Cross Section at 100-B/C Area, Hanford . . . . .	3-2
3-2	Location of the 116-C-1 Liquid Waste Trench in the 100-B/C Area, Hanford . . . . .	3-3
3-3	Location of the 100-D-1B Liquid and Sludge Waste Trench in the 100-D/DR Area, Hanford . . . . .	3-5
3-4	An Oblique Aerial View of the 116-C-1 Waste Trench and the Sampling Locations . . . . .	3-7
3-5	An Aerial View of the 100-D-1B Trench . . . . .	3-8
4-1	Particle-Size Distribution for 116-C-1 (Batch II), and 116-D-1B (Batch III) Soils . . . . .	4-6
4-2	A Scanning Electron Micrograph (Back-Scattered Mode) of a Plagioclase Feldspar Particle. The bright inclusions are hornblende, and the darker areas are quartz inclusions . . . . .	4-16
4-3	A Scanning Electron Micrograph (Back-Scattered Mode) of a Sodium Feldspar Fragment with Potassium Feldspar Inclusions (Lighter Areas) . . . . .	4-16
4-4	A Scanning Electron Micrograph of a Plagioclase Feldspar Grain with Inclusions of Hornblende (Light Areas), and Fe-Titanates (Very Bright Regions) . . . . .	4-17
4-5	Optical Micrograph of Soil Particles With White Coatings and Reddish-Yellow Stains . . . . .	4-17
4-6	A Scanning Electron Micrographic of Kaolinite Coatings Removed from the Sand-Sized Particles from 116-D-1B Soil . . . . .	4-18
4-7	Autoradiograph of Radionuclide Located on Mica Edge . . . . .	4-18

9443221.0012

## Figures (cont.)

5-1	Gilson Wet-Vac Wet-Sieving Unit . . . . .	5-2
5-2	Mass and Radionuclide Distribution in 116-C-1 (Batch I) Soil . . . . .	5-4
5-3	Mass and Radionuclide Distribution in 116-D-1B (Batch III) Soil . . . . .	5-6
5-4	Residual Radionuclide Activity in Composite Fraction of 116-D-1B Soil as a Function of Fractional Reduction of Radionuclide Activity in 2- to 0.25-mm-Sized Fraction . . . . .	5-7
6-1	Laboratory-Scale Attrition Scrubber with Servo-Controlled Motor . . . . .	6-2
6-2	A Close-Up View of the Opposing Pitch Impellers . . . . .	6-2
6-3	Percentage Reduction in Radionuclide Activity as a Function of Pulp Density . . . . .	6-5
6-4	Percentage Reduction in Radionuclide Activity as a Function of Input Energy . . . . .	6-7
6-5	Percentage Fines Generated During Attrition Scrubbing as a Function of Input Energy . . . . .	6-8
6-6	Observed Functional Relationships Between Impeller Torque and Time During Attrition Scrubbing Tests . . . . .	6-9
6-7	Optical Micrograph of Attritted Soil Particles from 116-D-1B Soil . . . . .	6-10
6-8	Scanning Electron Micrograph of a Mica Particle from 116-D-1B Soil . . . . .	6-10
6-9	Effects of Attrition Scrubbing on Equivalent Particle Diameters . . . . .	6-11
6-10	Effects of Attrition Scrubbing on Roundness Factor of Soil Particles . . . . .	6-12
7-1	Laboratory-Scale Rod Mill Used for Autogenous Grinding . . . . .	7-1
11-1	A Soil-Washing Scheme for 116-D-1B Trench Soils . . . . .	11-8
11-2	Process Flow Diagram for Pilot-Scale Tests on 116-D-1B (Batch III) Trench Soil . . . . .	11-10



## Tables

2-1	Soil-Washing Feasibility Tests of Radionuclide-Contaminated Soils . . . . .	2-2
3-1	Average Activities of Radionuclides in Soils from 116-C-1 and 116-D-1B Trenches . . . . .	3-4
4-1	Particle-Size Distribution Data for 116-C-1 Batch II and 116-D-1B Batch III Trench Soils . . . . .	4-7
4-2	Specific Gravity of 116-C-1 and 116-D-1B Trench Soils . . . . .	4-7
4-3	Total Organic Carbon Content . . . . .	4-7
4-4	pH Measurements . . . . .	4-8
4-5	Cation Exchangeable Capacity of 100 Area Soil Samples . . . . .	4-8
4-6	Major Element Concentrations (%) in 100 Area Soil Samples . . . . .	4-9
4-7	Trace Element Concentrations (mg/kg) in 100 Area Soil Samples . . . . .	4-10
4-8	Radionuclide Data for 100 Area Soil Samples . . . . .	4-11
4-9	Activities and Concentrations of Contaminant Radionuclides in 116-C-1 and 116-D-1B Trench Soils . . . . .	4-11
4-10	Analyses of Extracts from Toxicity Characteristics Leaching Procedure . . . . .	4-12
4-11	Sequential Extraction Data for 116-C-1 and 116-D-1B Trench Soils . . . . .	4-15
4-12	Mineralogy of 116-D-1B Trench Soil . . . . .	4-15
4-13	Types of Contaminant-Mineral Associations . . . . .	4-15
5-1	Contaminant Radionuclide and Mass Distribution Data for 116-C-1 Batch I and 116-D-1B Batch III Soils . . . . .	5-5
6-1	Attrition Scrubbing Data for 2- to 0.25-mm-Sized Fractions of 116-D-1B Soil: Relationship Between Pulp Density and Reduction in Radionuclide Activity . . . . .	6-4

6-2	Results from Attrition Scrubbing Tests on 116-D-1B Soil Sample: Reduction in Contaminant Radionuclide Activity as a Function of Input Energy . . . . .	6-6
6-3	Results of Optimized Attrition Scrubbing Tests with Water and an Electrolyte on 2- to 0.25-mm-Sized Fractions of 116-D-1B Soil . . . . .	6-13
7-1	Autogenous Grinding Data for Gravel Fractions from 116-C-1 Soil . . . . .	7-3
8-1	Standard Chemical Extraction Data for 116-D-1B Soil . . . . .	8-3
8-2	Specific Chemical Extraction Data for 116-D-1B Soil . . . . .	8-3
8-3	Effects of Specific Extractant Concentration and Solid/Extractant Ratio on Contaminant Radionuclide Removal from 116-D-1B Soil . . . . .	8-4
8-4	Specific Chemical Leaching Data for Gravel-Sized Fractions from 116-C-1 Soil . . . . .	8-5
9-1	Results from Two-Stage Attrition Scrubbing in Water and Electrolyte Media Conducted on Sand Fractions from 116-D-1B Soil . . . . .	9-4
9-2	Results from Autogenous Grinding with an Electrolyte and a Specific Extractant Conducted on Gravel-Sized Fractions from 116-C-1 Soil . . . . .	9-5
10-1	Results of Flocculation Experiments on Waste Waters from 116-D-1B (Batch III) Soil-Washing Tests . . . . .	10-4
11-1	Wet-Sieving Processing Option for 116-D-1B (Batch III) Soil . . . . .	11-5
11-2	Wet-Sieving and Attrition Scrubbing Options for 116-D-1B (Batch III) Soil . . . . .	11-6

## 1.0 INTRODUCTION

The 100 Area of the Hanford Site contains nine inactive nuclear reactors that were operated for the production of fissionable material (Figure 1-1). All these water-cooled reactors (B, C, D, DR, F, H, KE, KW, and N) situated along the southern bank of the Columbia River have been shut down and currently are being evaluated for decommissioning. Waste streams that were generated during the operation of these reactors were disposed of in the vicinity, resulting in wide-spread contamination of both soil and water. The bulk of the contamination originated from the disposal of very large volumes of reactor cooling water containing both fission and activation products.

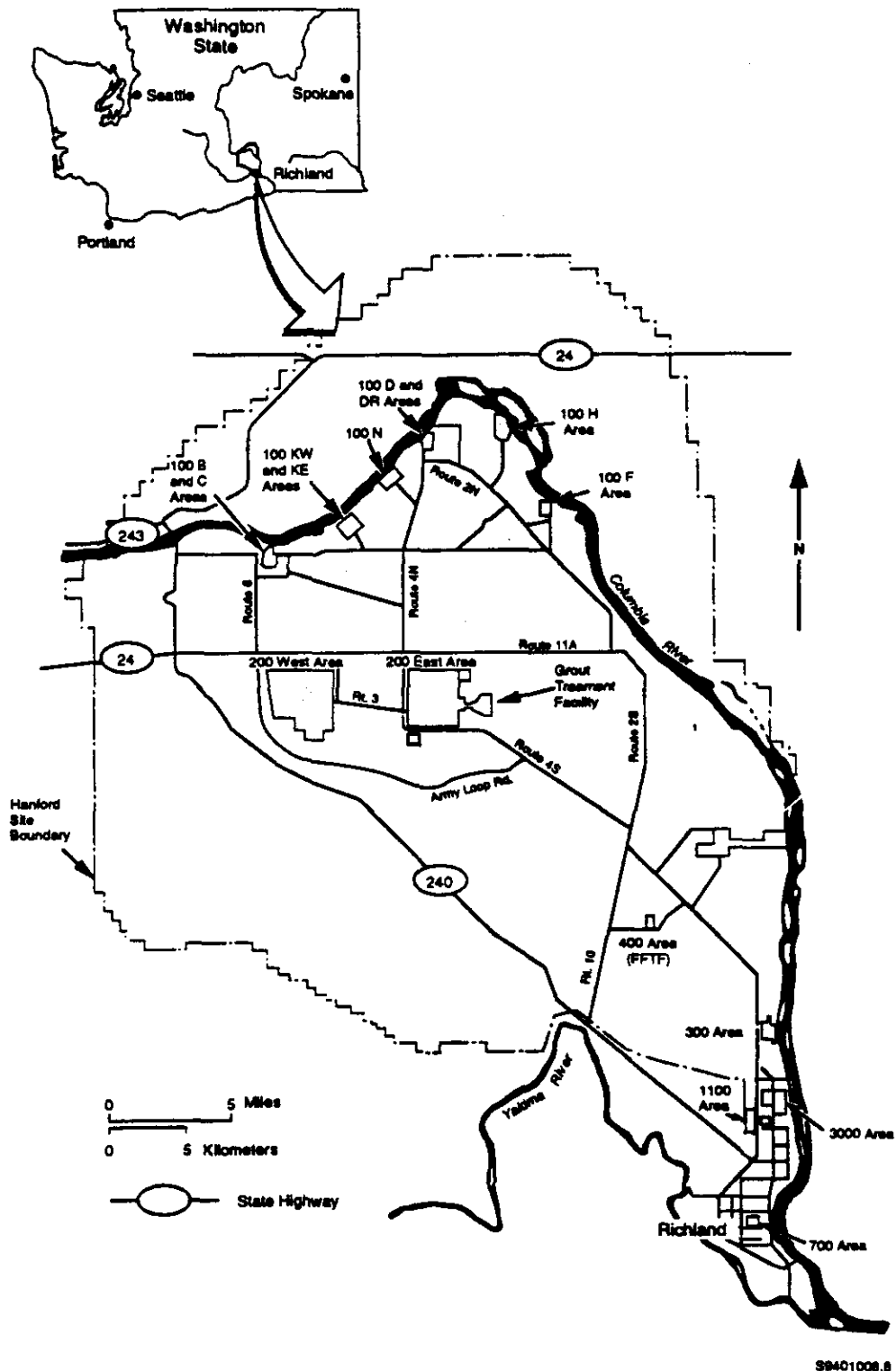
The contaminated cooling water was discharged into engineered trenches and cribs that resulted in substantial volumes of soil being contaminated with radionuclides. Additionally, leaks in the cooling water transfer systems also contaminated the soil and groundwater. The extent of the contamination resulted in the inclusion of the 100 Areas of the Hanford Site in the National Priorities List (NPL) compiled under the *Comprehensive Environmental Response, Compensation, and Liability Act* (CERCLA) by the U.S. Environmental Protection Agency (EPA).

The types and the extent of environmental contamination in the 100 Areas have been investigated as part of Remedial Investigation and Feasibility Studies (RI/FS) (DOE-RL 1992a). Based on these data, several remedial measures have been examined and documented in a report (DOE-RL 1992d). Because of the large volumes of contaminated soils that need remediation, any treatment technology that reduces the volume of soil to be disposed of as waste can offer significant savings in remedial costs. One of the more promising remedial measures is soil-washing. Therefore, soil-washing treatability studies were conducted on the 100 Area soils to derive specific data for evaluating the applicability of this technology.

A test plan was developed (DOE-RL 1992b) to conduct laboratory-scale tests on two representative 100 Area soils contaminated during cooling water disposal. These laboratory-scale bench tests were designed to fulfill the initial screening phase of an overall three-phase treatability study. The objective of this treatability study was to evaluate the use of physical separation systems and chemical extraction methods as a means of separating chemically and radioactively contaminated soil fractions from uncontaminated soil fractions. The other two phases of the treatability study, namely, remedy selection and design, rely on the data obtained during the screening phase, which is the subject of this report.

The laboratory-scale screening tests to be performed on 100-Area soils were described as a set of procedures (Freeman et al. 1993). These procedures include physical, chemical, and mineralogical characterization of the soils; soil fractionation through wet-sieving to characterize particle mass and contaminant distribution; contaminant mobilization through physical (attrition scrubbing and autogenous grinding) and chemical treatments (extraction and leaching); and the treatment of wash water and spent extractants.

Figure 1-1. The Location of the Hanford Site.



## 2.0 SOIL-WASHING TECHNOLOGY

### 2.1 DESCRIPTION OF THE TECHNOLOGY

Soil washing is a remediation technology aimed at removing inorganic, organic, and radionuclide contaminants to the extent that the residual levels of these contaminants in coarse fraction (the bulk) of the treated soil are below selected safe levels. The remediated coarse soil is returned to the excavation source, and the smaller volume of soil containing the contaminants is disposed. Typically, soil washing is accomplished with physical or chemical techniques, or a combination.

The effectiveness of soil washing as a remediation technique depends mainly on the type of binding processes that exists between the contaminants and the soil particles. Contaminants in soils may be found adsorbed or precipitated on the soil mineral surfaces or as distinct particles. If a contaminant exists mainly in the adsorbed form, the finer soil particle fractions tend to contain higher concentrations of that contaminant. This phenomenon can be attributed to the fact that finer particles possess greater surface areas per unit mass than the coarser sized fractions of soils. If the contaminants exist mainly as coatings or precipitates on soil particles, washing with water, physical agitation through attrition, may dislodge and separate the contaminated fine material, or the precipitates may be dissolved by using chemical extractants. Contaminants that exist as distinct particles can also be separated if they possess significantly different specific gravities than the bulk minerals in a soil matrix.

Typically, the initial step in the soil-washing process consists of separating the soils into different sized fractions. This is generally accomplished by using hydraulic classification techniques such as screening or hydrocyclones. If necessary, hydraulic jigs may also be used to separate contaminant-bearing particles based on their differences in size and specific gravity. Depending on the soil and the types of contaminants, this initial physical separation process may accomplish the goal of recovering clean soil that constitutes the major fraction of the soil mass and isolating the major fractions of the contaminants in a smaller volume of fine-grained soil that can removed for disposal.

If the contaminant distribution among various particle fractions of a soil indicates that the levels of contamination in the coarser soil fractions exceed designated safe levels, additional physical and/or chemical treatments may be necessary to release the surface-bound contamination. The principal physical method used to release the contaminants, through grinding action, is attrition scrubbing. In this method, the moist soil particles are agitated vigorously and made to attrit against each other to dislodge the surface-bound contaminants. Such scrubbing will generate contaminant-laden fine material that is washed out of coarser material containing significantly lower concentrations of contaminants. Attrition scrubbing may also be conducted with surfactants or electrolytes to attenuate the re-adsorption of released contaminants onto the freshly exposed surfaces of the soil particles. Typically, the electrolytes may either be acidic or alkaline solutions that enhance the solubility of contaminants and may also contain ligands that chelate mobilized cationic contaminants.

Additionally, the electrolytes may include ionic components that reduce readsorption of released contaminants by blocking the adsorption sites on soil particle surfaces. Generally, attrition scrubbing with a suitable electrolyte increases contaminant removal through a synergistic combination of physical and chemical processes. Chemical treatments used to remove contaminants from coarse soil fractions typically consist of an extractive process that uses solvents (aqueous acids, alkalis, organic solvents) to desorb and dissolve the contaminant-bearing particles and coatings from the soils.

## 2.2 FEASIBILITY OF SOIL-WASHING RADIONUCLIDE-CONTAMINATED SOILS

A literature review indicated that a number of bench-scale tests have been conducted to assess the feasibility of using soil-washing as a technique to remediate soils contaminated with various radionuclides (Table 2-1). A number of these studies were focused on the feasibility of soil washing to remediate soils contaminated with  $^{238/239}\text{Pu}$  and  $^{241}\text{Am}$ . The effects of scrubbing action on soil washing were studied by Horton and Albenesius (1976). They conducted tests on samples of plutonium-contaminated soils from the Savannah River Laboratory site in Georgia by agitating these soils in a flow-through container system. Their data indicated that increasing agitation time resulted in decreasing levels of Pu in the sand fraction with concomitant increase in the amount of contaminated silt-clay fraction.

Table 2-1. Soil-Washing Feasibility Tests of Radionuclide-Contaminated Soils.

Soil Source	Contaminant Radionuclide	References
Savannah River site, Georgia	$^{238}\text{Pu}$	Horten and Albenesius (1976)
Rocky Flats site, Colorado	$^{238}\text{Pu}$ , $^{241}\text{Am}$	Navratil and Kochen (1982) Stevens and Rutherford (1982)
Rocky Flats, Colorado; Hanford, Washington; Mound, Ohio; Idaho Falls, Idaho; Los Alamos, New Mexico	$^{238,239}\text{Pu}$ , $^{241}\text{Am}$	Stevens et al. (1982)
Montclair/Glen Ridge, New Jersey	$^{226}\text{Ra}$ , $^{230}\text{Th}$	Richardson et al. (1989) Phillips et al. (1993)
Idaho Falls, Idaho	$^{60}\text{Co}$ , $^{137}\text{Cs}$	Gombert (1992); Murray (1993)
Fernald, Ohio	U	Soil Decon Task Group (1993)

Navratil and Kochen (1982) investigated the effectiveness of wet- and dry-screening, attrition scrubbing, and chemical extraction for releasing  $^{238}\text{Pu}$  and  $^{241}\text{Am}$  from contaminated soils (total activity: 900 to 140,000 pCi/g) from the Rocky Flats Plant site in Colorado.

The results showed that dry-screening was not as effective as wet-screening for isolating the radionuclides in the fine fraction. Using wet-screening, about 65 % by mass of the soil (>2.4-mm in size) containing low activities of these radionuclides (<6 pCi/g of Pu and <3 pCi/g of Am) was recovered. Additionally, using attrition scrubbing of the 2- to 0.42-mm soil fraction, the activity of Pu in this fraction was reduced from 300 pCi/g to <20 pCi/g. Among the chemical extractants tested, Calgon, oxalic acid, and a proprietary detergent removed major fractions (98 % to 99 %) of plutonium and americium from the 4- to 0.42-mm sized fraction. Another study was conducted by Stevens and Rutherford (1982) on soils from the same site. They examined the effects of more intense agitation on chemical extractability of these radionuclides. The data indicated that the extractability of the contaminants was significantly increased by a combination of intense agitation and dissolution.

The effects of repetitive chemical extraction on the mobilization of plutonium and americium from contaminated soils from five different sites were investigated by Stevens et al. (1982). In this study, soils were successively extracted for a total of five times with three different extractants (sodium hydroxide, 2N hydrochloric acid, and a mixed extractant consisting of 2 % nitric acid, 0.2 % hydrofluoric acid, 2 % pine oil, and 5 % Calgon solution). The data showed that these three extractants differed in their effectiveness for removing the contaminants from each type of soil. For instance, hydrochloric acid was the most effective extractant for releasing the contaminants from the soils sampled from sites at Hanford, Idaho, and Mound, whereas the sodium hydroxide was the most effective extractant for the Rocky Flats soil. The mixed extractant was most effective for releasing plutonium and americium for the Los Alamos soil.

A bench-scale soil-washing study consisting of wet-sieving was conducted by Richardson et al. (1989) on soils contaminated with radium and thorium from a site in Montclair/Glen Ridge, New Jersey. The results of these tests indicated that up to 56 % of the mass (>74  $\mu\text{m}$  in size) of soil could be recovered with an activity of 16 pCi/g. A recent pilot-scale study was conducted on the same soil by Phillips et al. (1993). The data from this test showed that at least 54 % of the soil mass (>74  $\mu\text{m}$ ) containing an activity of <12 pCi/g could be recovered as remediated material and returned to the excavation site.

Recently, the extractability of  $^{60}\text{Co}$  and  $^{137}\text{Cs}$  from contaminated soil from a waste pond in Idaho Falls, Idaho, was tested by Gombert (1992). This study consisted of using a sequence of different chemical extractants to assess the release of these two radionuclides from the finer fraction (<0.425 mm) of the contaminated soil. The results indicated that up to 84 % of cobalt could be extracted from the soil. By contrast, only a minor fraction (about 20 %) of cesium was extractable by this procedure. The same waste pond soils were used in a more extensive bench-scale soil-washing feasibility study conducted by Murray (1993). This test included a number of soil-washing unit operations such as wet-sieving, attrition scrubbing, froth flotation, and chemical extraction. The effects of a number of parameters related to these processes (temperature, pulp density, retention time, the types and concentrations of extractants) were also tested. The data from these tests showed that using a soil-washing system consisting of wet-sieving, froth flotation, multiple-stage attrition scrubbing, and chemical extraction resulted in recovering about 92 % of the soil mass with a residual cesium activity below the target performance level of 690 pCi/g.

A set of soil-washing studies was conducted by the Soil Decon Task Group (1993) to check the feasibility of removing U from two contaminated soils from Fernald, Ohio. The data indicated that a number of chemical extractants such as carbonate solutions, citric acid, Tiron with sodium dithionite solutions, and a mixture of sodium salts of citrate, bicarbonate, and dithionite effectively removed substantial amounts of uranium from these soils. Attrition scrubbing combined with chemical extractants (sodium and ammonium carbonate, citric and sulfuric acids) also released significant amounts of uranium from these soils.

### 2.3 TARGET PERFORMANCE LEVELS FOR RADIONUCLIDES

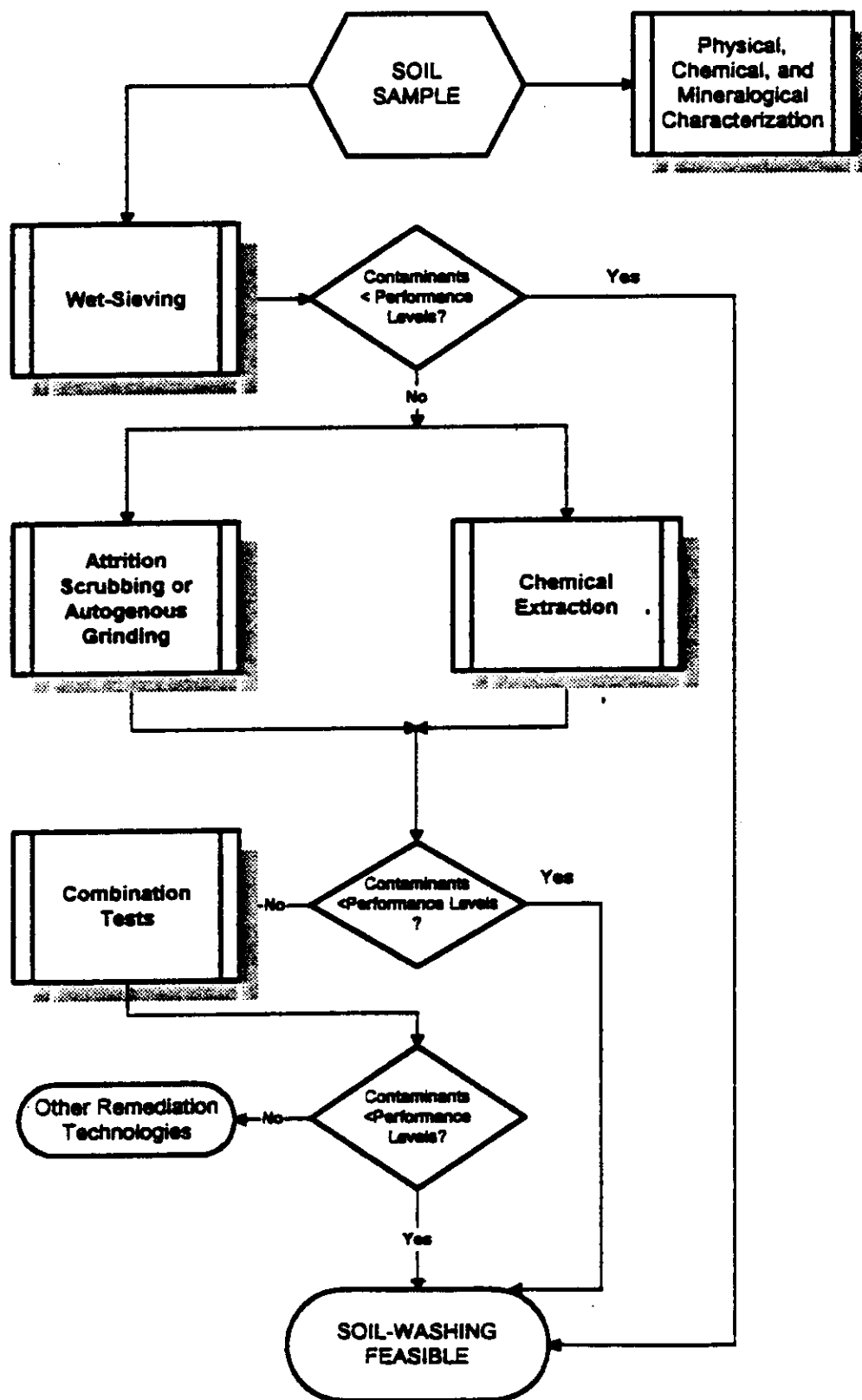
The test plan for 100 Area soil-washing feasibility tests (DOE/RL, 1992b) included a set of target performance levels (TPL) for various radionuclides. According to these criteria, treated soils were to be considered clean if the residual activities of radionuclides were found to be below these specified TPL. Recently, based on effective dose equivalents of 10 mrem/yr a revised set of TPL values were established. (See Table 6-2 in WHC, 1988). Therefore, this new set of TPL values will be used to assess the level of soil contamination and to infer the effectiveness of soil-washing processes.

### 2.4 A SOIL-WASHING TEST SCHEME FOR 100 AREA SOILS

A review of published data from a number of laboratory-scale soil-washing tests on radionuclide-contaminated soils indicated that a test scheme for soil-washing could be developed by combining a number of unit operations. Therefore, a scheme was formulated for testing the feasibility of soil-washing 100 Area waste trench soils (Figure 2-1). This scheme included, as an initial step, physical, chemical, and mineralogical characterization. These characterization data are essential for selecting relevant treatment methods for each soil. The data regarding the mass distribution of particle sizes and the activities of contaminant radionuclides in these soil fractions were obtained from wet-sieving data. If the residual radionuclide activities of a substantial portion of the bulk soil was found to be less than the specified target performance levels (TPL), then wet-sieving alone could be used to remediate that soil. By contrast, if the wet-sieving data indicate that the coarser sized fractions contain residual activities that exceed the TPL, additional physical (attrition scrubbing or autogenous grinding) and chemical (extraction or leaching) treatment tests were conducted. Remediation would be feasible if these treatments reduced the radionuclide activities in coarser fractions to below the TPL. If this goal was not accomplished, additional tests that combine both physical and chemical treatments were conducted. These combination tests typically included multi-stage attrition scrubbing or autogenous grinding with electrolytes or chemical extractants. Soil-washing would be considered effective for a soil if the residual radionuclide activities in the treated coarser soil fractions were found to be less than the TPL.



Figure 2-1. A Scheme for Soil-Washing Feasibility Tests on 100 Area Soils.



### 3.0 SOIL SAMPLES FROM THE 100 AREA

#### 3.1 SITE DESCRIPTION

The 100 Area of the Hanford Site is located in a structural basin (Pasco basin) on the Columbia Plateau. A generalized geologic cross section through the 100-B/C Area (Figure 3-1) shows a sequence of unconsolidated surficial sedimentary deposits (Hanford formation). The topmost layer of this formation consists of a thin layer (1 to 15 ft thick) of light brown, fine, slightly silty, eolian deposits.

The principal sediments of the Hanford formation underlying the surficial layer range from twenty to several hundred feet in thickness and consist of poorly sorted, unconsolidated glaciofluvial material (Pasco gravels). The origin of these sedimentary deposits is believed to be the periodic cataclysmic floods that occurred during the late Pleistocene epoch (Baker 1981; Mullineaux et al. 1977). The Pasco gravels are variable mixtures of particle sizes that range from boulders to silt. The bulk of Pasco gravels are very coarse-textured and are classified as silty sandy gravels typically consisting of about 50% gravel, 40% sand, and 10% silt (DOE-RL 1988). Mineralogically, these deposits consist of quartz, feldspars, and ferromagnesian minerals (DOE-RL 1992c). All the trenches and cribs that were engineered for waste disposal in the 100 Area were located in the Pasco gravels.

The Ringold Formation, ranging up to several hundred feet in thickness, underlies the Hanford formation. This formation, late Miocene to late Pleistocene in age, consists of stratified deposits of material ranging in size from gravel to clay. The uppermost unconfined aquifer system in the Hanford area, ranging up to 300 ft in depth, is located within the Hanford and Ringold Formations. The Saddle Mountain Basalt Formation underlies the stratified Ringold deposits.

#### 3.2 TEST SITES

Two waste disposal trenches were selected for soil-washing feasibility tests because these sites are representative of a number of similar sites in the 100 Areas, and these trenches contain a number of contaminants (DOE-RL 1992b). The 116-C-1 liquid waste trench is located in the 100-BC-1 Operable Unit of the 100 Areas (Figure 3-2). This unlined trench is 500 ft long, 50 ft wide, and 16 ft deep. This trench received reactor cooling effluent from the 116-C-5 retention basin from 1952 to 1958. During this period, this trench received a total estimated volume of 26 million gal of effluent that was contaminated with activation and fission products caused by fuel-cladding failures in the 100-C Reactor. The 116-C-1 trench also received Cr-bearing wastes in the form of sodium dichromate (DOE-RL 1992b). This trench was partially filled with rocks to prevent the spread of contamination by wind action. Based on the soil sampling conducted in 1976, the total volume of contaminated soil in this trench was estimated by Dorian and Richards (1978) to be about 2.7 million ft<sup>3</sup> (600 by 150 by 30 ft). The average activities of

Figure 3-1. Generalized Geologic Cross Section at 100-B/C Area, Hanford.

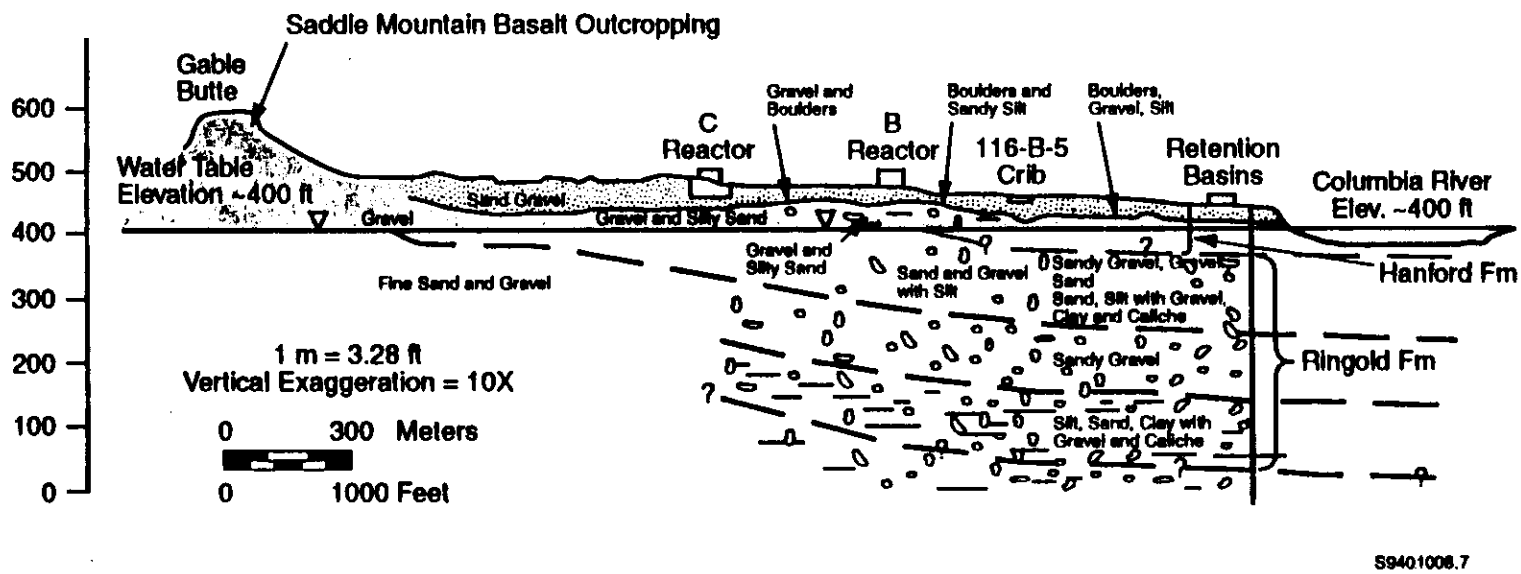
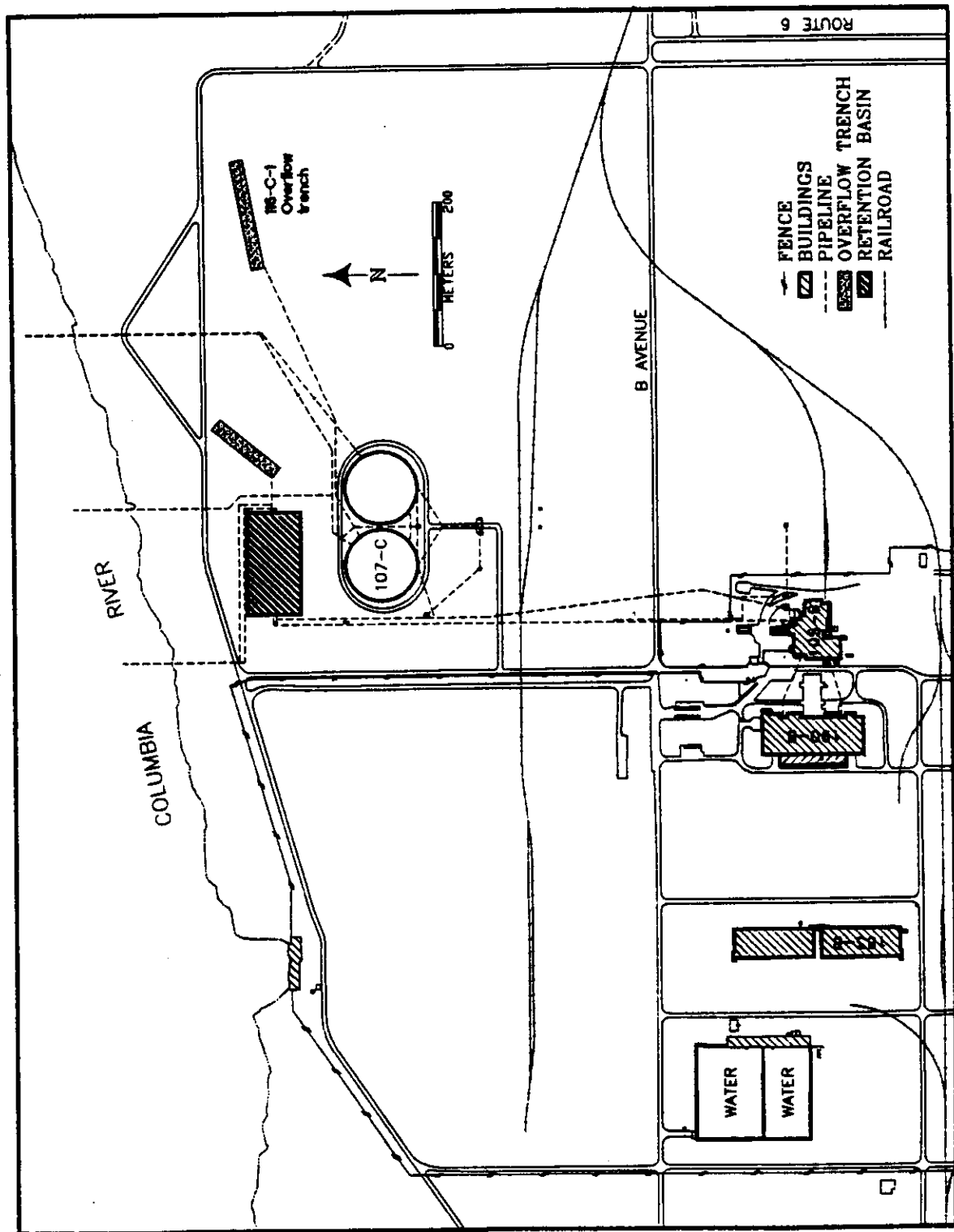


Figure 3-2. Location of the 116-C-Liquid Waste Trench in the 100-B/C Area, Hanford.



radionuclides and an inventory of total activity for the 116-C-1 trench (Dorian and Richards 1978) are listed in Table 3-1. These data indicate the soil in this trench may contain activity levels of  $^{60}\text{Co}$ ,  $^{137}\text{Cs}$ ,  $^{152}\text{Eu}$ , and  $^{154}\text{Eu}$  that exceed the proposed target performance levels (TPL).

Table 3-1. Average Activities of Radionuclides in Soils from 116-C-1 and 116-D-1B Trenches\*.

Radionuclide	116-C-1 Average (pCi/g)	116-D-1B Average (pCi/g)	Accessible Soil Activity Limits** (pCi/g)
$^{60}\text{Co}$	180	14	7.1
$^{134}\text{Cs}$	2.2	0.35	10
$^{137}\text{Cs}$	39	44	30
$^{152}\text{Eu}$	130	31	15
$^{154}\text{Eu}$	67	5.9	14
$^{155}\text{Eu}$	6.8	63	630
$^{90}\text{Sr}$	11	14	2800
$^{235}\text{U}$	0.2***	0.18***	170
$^{238}\text{U}$	***	***	370
$^{239/240}\text{Pu}$	0.74	0.48	190
Contaminated Soil Volume ( $\text{ft}^3$ )	$2.7 \times 10^6$	$2.1 \times 10^5$	--
Contaminated Soil Mass (g)	$1.8 \times 10^{11}$	$1.4 \times 10^{10}$	--
Total Activity (Ci)	79	2.6	--

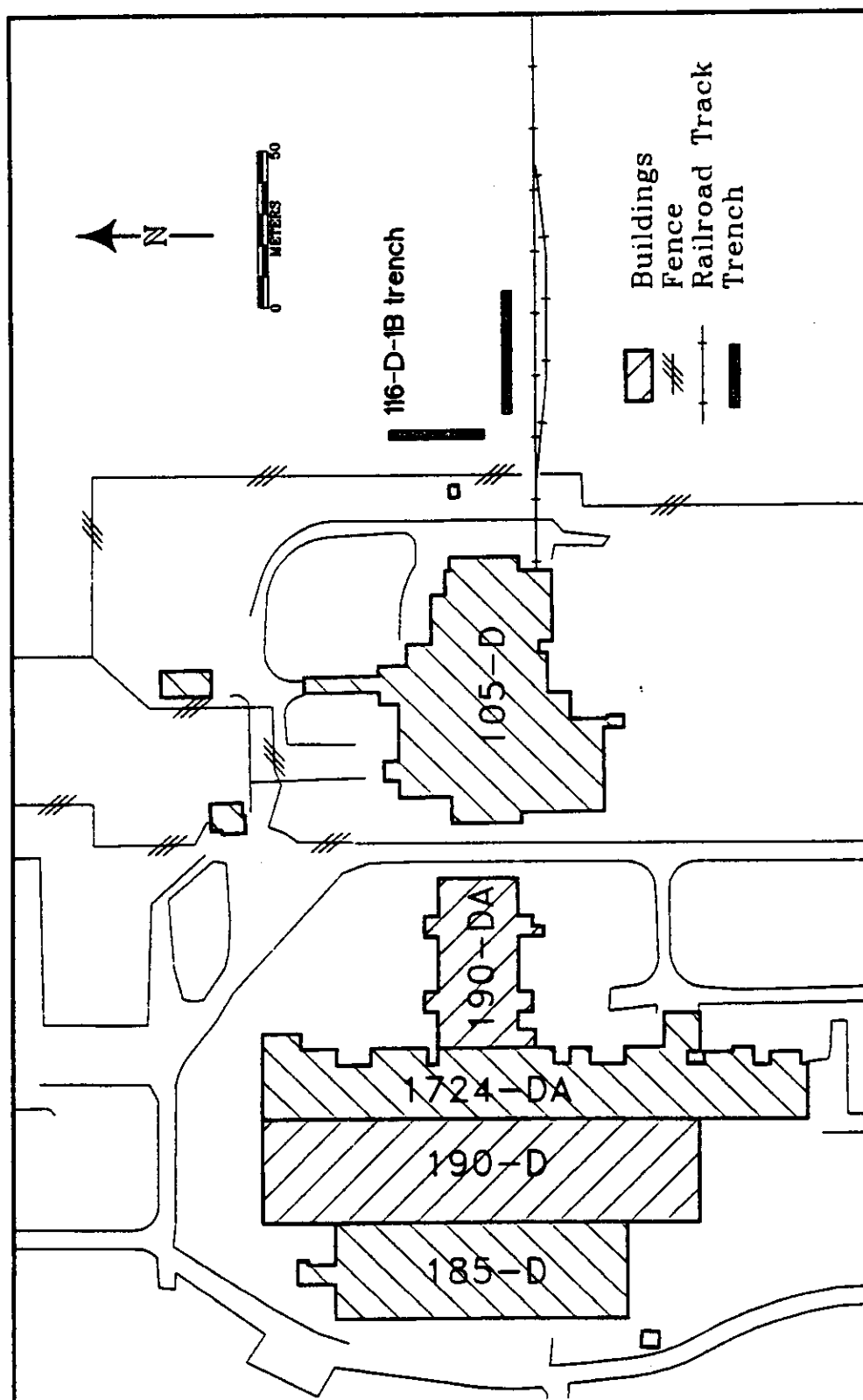
\*Source: Dorian and Richards (1978).

\*\* Table 6-2, WHC (1988).

\*\*\* Measured as total U.

The 116-D-1B liquid waste trench is located in the 100-DR-1 Operable Unit of the 100 Areas (Figure 3-3). This trench is 100 ft long, 10 ft wide, and 15 ft deep. It was used mainly to dispose of radionuclide-contaminated sludge and water from the 118-D-6 fuel storage basin. This material originated as the result of storing ruptured fuel elements in the 118-D-6 basin. The 116-D-1B trench was used from 1953 through 1967 and probably received a major fraction of sludge (estimated to be 110,000 lb) that originated in the fuel

Figure 3-3. Location of the 100-D-1B Liquid and Sludge Waste Trench in the 100-D/DR Area, Hanford.



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storage basin. Other contaminants disposed of in this trench included about 1540 lb of sodium dichromate, 4400 lb of either sodium formate or oxalate, and 4400 lb of sodium sulfamate. After waste discharges were stopped in 1967, the trench was blanketed with a layer of clean soil. As a result of sampling and analyses of soils from this trench, the total volume of contaminated soil was estimated by Dorian and Richards (1978) to be about 210,000 ft<sup>3</sup> (150 by 40 by 35 ft). The average activities of radionuclides and an inventory of total activity for this trench (Dorian and Richards 1978) are listed in Table 3.1. These data indicate the soil in this trench may contain radionuclides <sup>60</sup>Co, <sup>137</sup>Cs, and <sup>152</sup>Eu at activities that exceed the TPL.

### 3.3 SOIL SAMPLE COLLECTION

All the soil samples were collected by WHC personnel and transported to the laboratory in Department of Transportation (DOT)-specified containers. The following description of the sampling operations is based on the field log data recorded by Guzek and Field (1993). The first sample was collected at a depth of 20 ft from a pit excavated in the middle of the 116-C-1 trench (Figure 3-4). The sample was collected in 11 containers of 5-gal capacity each and transported to the laboratory. This sample was designated as "Batch I." Field monitoring of the soil samples with a handheld Geiger-Mueller probe indicated the activity ranged between 300 to 500 counts per minute (cpm). A week after the first sampling was completed, a second set of soil samples (Batch II) was obtained from the vicinity of the inlet pipe.

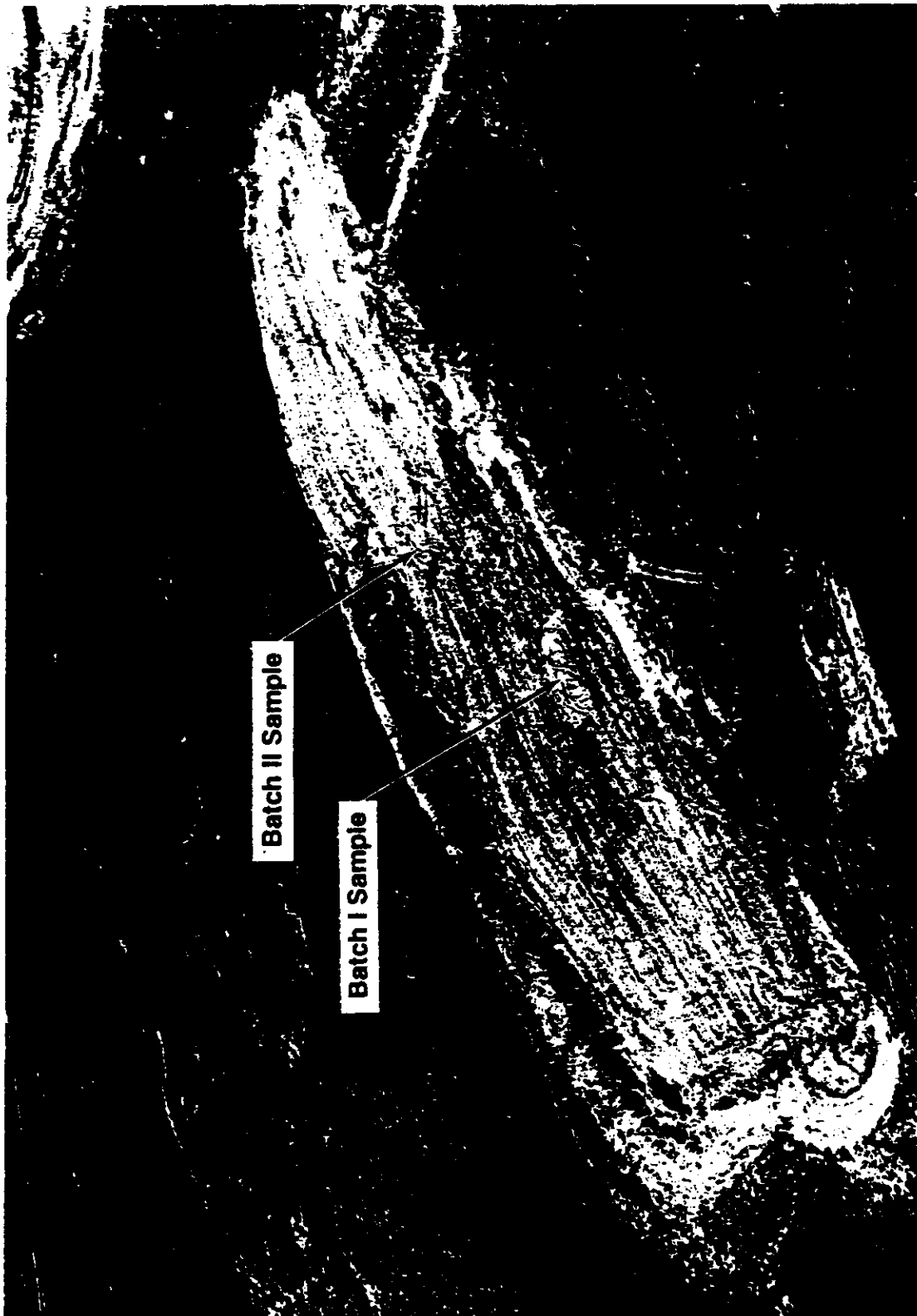
Field monitoring indicated radioactivity increasing from approximately 1000 cpm at a 10-ft depth to about 20,000 cpm at a depth of 20 ft. At a depth of 22 to 24 ft, the measured activities were lower (1500 to 2000 cpm), indicating that the maximum soil contamination in this trench occurs near the inlet at a depth of 20 ft. The soil samples were collected from 10 ft depth, at 15 to 18 ft and at 18 to 20 ft depth intervals. A total volume of 55 gal of soil was collected and transported to the laboratory.

The soil samples (Batch III) from the 116-D-1B (see Figure 3-5) trench were collected at depth intervals of 5 to 10 ft, and 15 to 20 ft. Activities at these sampling depths were about 600 cpm. The activity was observed to decline to a level of 150 to 200 cpm at a depth of approximately 22 to 24 ft. A total volume of 55 gal of soil was collected and transported to the laboratory in 11 5-gal containers.

### 3.4 SOIL SAMPLE PREPARATION

The soil samples received in 5-gal capacity containers were spread on drying trays placed in fume hoods and, as recommended by the ASTM standard practice (D 421-85), thoroughly air-dried. All air-dried samples were friable, and therefore easily disaggregated. Following disaggregation, each soil sample was dry-screened using a 13.5-mm screen to isolate gravel-sized material and a 2-mm screen to separate material finer than coarse sand.

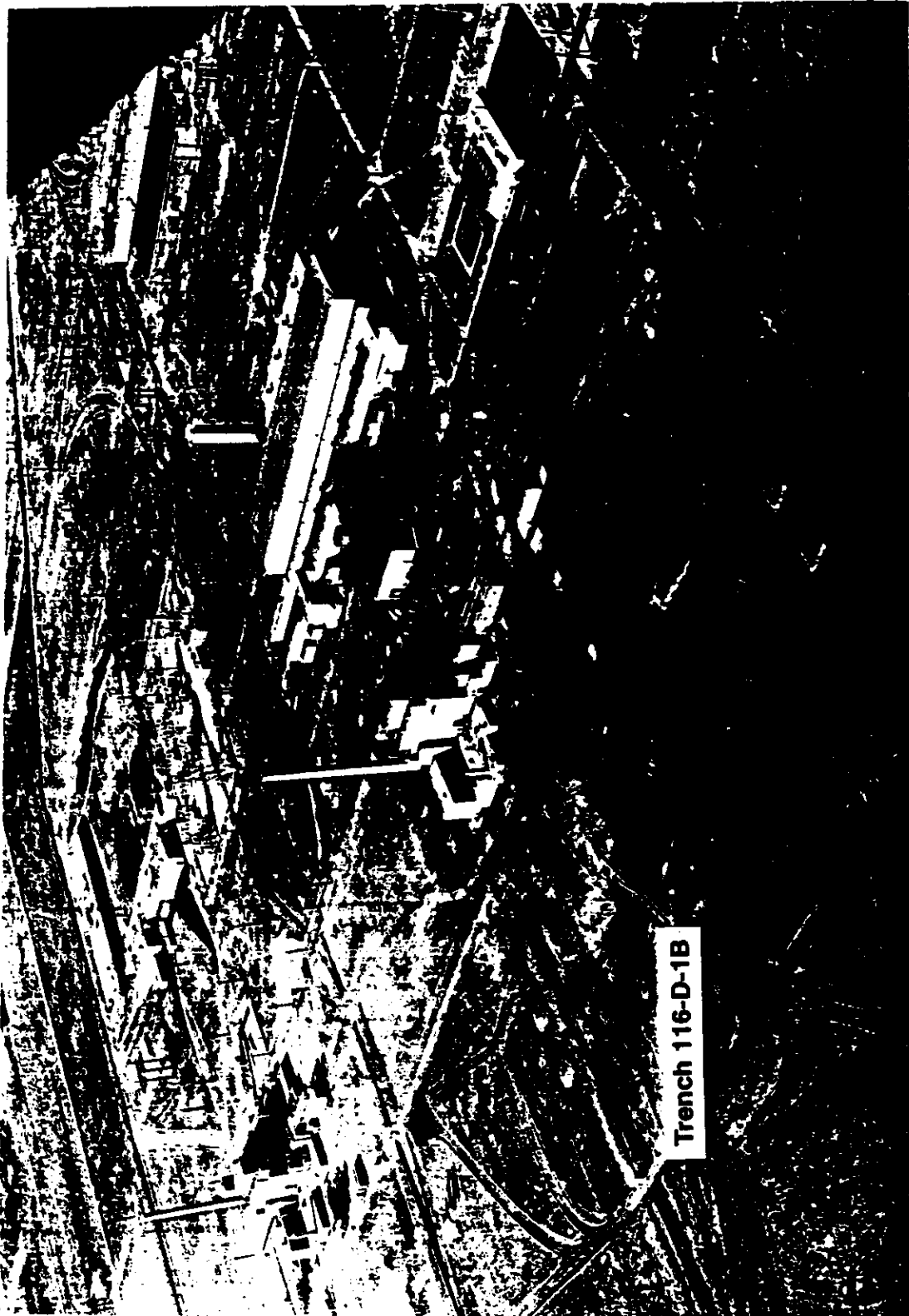
Figure 3-4. An Oblique Aerial View of the 116-C-1 Waste Trench and the Sampling Locations.



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Figure 3-5. An Aerial View of the 100-D-1B Trench.



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Subsamples of air-dried <2-mm material were obtained by homogenizing, coning, and quartering the soil (ASTM C 702-87). Because of the large volume of the sample, subsampling was accomplished by compositing air-dried and dry-screened soil sample into four equal batches. Next, each of these four batches was coned and quartered, and one randomly selected quarter from each of the four batches was composited into a single batch. This single batch was stored, and representative subsamples were drawn from it for subsequent work. If this single batch was used up during testing, additional batches were prepared by compositing the remaining soil into an appropriate number of batches and repeating the process of coning and quartering until a new single batch of soil was obtained.

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## 4.0 CHARACTERIZATION OF SOILS

The principal objectives of soil characterization was to determine the properties (physical, chemical, and mineralogical) that govern the contaminant partitioning and release behavior of soils during the washing process. Typically, all the characterization tests can be conducted on <2-mm size fractions. However, there are no test protocols to measure characteristics such as, Total Organic Carbon (TOC), pH, Cation Exchange Capacity (CEC), and Toxicity Characteristics Leaching Procedure (TCLP) of gravel and cobble fractions. Only a limited number of tests such as, the particle size distribution, specific gravity, and radionuclide analysis can be conducted on very coarse (gravel and cobble) soil fractions. Therefore, this study included a complete suite of specified characterization tests for <2-mm size soil fractions, and only a limited number of applicable tests for the gravel fractions of 100 Area soils.

### 4.1 PHYSICAL CHARACTERIZATION

#### 4.1.1 Moisture Content

Gravimetric water contents of the soil samples were determined using a standard procedure (Gardner 1986). Air-dried soil samples (<2 mm) in tared containers were oven-dried at  $105 \pm 5^\circ\text{C}$  for 10 to 24 hours, cooled over a desiccant, and weighed. The gravimetric water content was computed as percentage change in soil weight before and after oven drying.

#### 4.1.2 Particle Size Distribution

The particle size of Batch II and Batch III soil samples was determined according to ASTM method D 422-63. According to this method, the distribution of particle sizes larger than 2 mm (retained on No. 10 sieve) is determined by dry-sieving. Soil fractions finer than 2 mm were dispersed, and the distribution of particles smaller than 0.075 mm was determined by measuring the sedimentation rate using a hydrometer. Following the hydrometer measurements, the soil sample was washed through a 0.075-mm (No. 200) sieve, and dried at  $110 \pm 5^\circ\text{C}$ , and material larger than 0.075 mm was dry-sieved through a set of sieves (Numbers 20, 40, 60, and 140). The weight percentage of soil finer than each specified size fraction was tabulated.

#### 4.1.3 Specific Gravity

The specific gravity of soil samples was determined according to the ASTM standard test methods. For soil particle fractions larger than 4.75 mm, ASTM method C 127-88 was used. The specific gravity of soil fractions finer than 4.75 mm was measured by ASTM method D 854-83. The specific gravity value of the whole sample was computed as the

weighted average of both soil fractions, as specified in ASTM D 854-83. Additionally, specific gravities of particles smaller than 2 mm were determined and used in calculating particle sizes by the hydrometer method (ASTM D 422-63).

## **4.2 CHEMICAL CHARACTERIZATION**

### **4.2.1 Total Organic Carbon**

The total organic carbon (TOC) contents of the soil samples were measured by the coulometric method (ASTM D 4129-88). In this method, soil-bound carbon is mobilized as carbon dioxide through combustion and acidification. The released carbon dioxide is absorbed into ethanolamine and measured by coulometric titration. The TOC values were calculated as percentage of the mass of soil.

### **4.2.2 Soil pH**

The pH of <2-mm sized fractions of all soils was determined by equilibrating soil samples with deionized distilled water (1:1) for 10 minutes and decanting, and measuring the pH of the supernatant with a calibrated glass electrode.

### **4.2.3 Cation Exchange Capacity**

Cation exchange capacity of the soils was determined according to the ammonium acetate method (Thomas 1986). According to this method, the exchangeable cations were displaced by equilibrating about 5 g of soil with 25 mL of 1N ammonium acetate solution for 30 min. After centrifuging the soil suspension, the supernatant was decanted and saved. The equilibration was repeated a second time, and the displaced supernatant was composited with the supernatant from the first equilibration. Finally, the combined supernatant was analyzed for the exchangeable cation content (Ba, Ca, Mg, Sr, and Na) by inductively coupled plasma-mass spectrometric analyses (ICP-MS) (PNL-ALO-280). Cation exchange capacity was calculated as the milliequivalent sum of all exchangeable cations per 100 g of soil.

### **4.2.4 Total Elemental Analyses**

The total elemental compositions of the soil samples (<2-mm-sized fractions of 116-C-1 Batch I and II, and 116-D-1B Batch III) were measured in duplicate by x-ray fluorescence spectrometry (PNL 7-40-48, Rev. 1). This method uses an iron target for analyzing Al, Ca, K, and Si; a Zr target for measuring As, Co, Cr, Cu, Fe, Pb, Mn, Ni, Rb, Se, Sr, Ti, and Zn; a Ag target for analyzing U; and a Gd target to determine Ag, Ba, Cd, Sb, and Zr.

#### 4.2.5 Radionuclide Analyses

The radionuclide contents of the three soil samples (<2-mm-sized fractions of 116-C-1 Batch I and Batch II, and 116-D-1B Batch III) were determined by specified standard procedures. Gamma ray spectrometry (PNL-ALO-106; PNL-ALO-464) was used to determine the activities of  $^{60}\text{Co}$ ,  $^{134}\text{Cs}$ ,  $^{137}\text{Cs}$ ,  $^{152}\text{Eu}$ ,  $^{154}\text{Eu}$ , and  $^{155}\text{Eu}$ . Analyses of  $^{90}\text{Sr}$  were conducted according to standard wet chemical separation and beta counting methods (PNL-ALO-106; PNL-ALO-463; PNL-ALO-465). The activities of Pu isotopes  $^{239/240}\text{Pu}$  were measured by acid digesting the samples (PNL-ALO-106), separating Pu (PNL-ALO-417; PNL-ALO-466), electroplating Pu on to counting disks (PNL-ALO-468; PNL-ALO-496), and counting by alpha spectrometry (PNL-ALO-469). Uranium activities in these samples were determined by fusing and dissolving the soil samples (PNL 7-40-78), and analyzing the solutions by ICP-MS (PNL-ALO-280; PNL-ALO-282).

#### 4.2.6 Toxicity Characteristic Leaching Procedure (TCLP)

The TCLP tests of 116-C-1 Batch II and 116-D-1B Batch III trench soils (<2-mm-sized fraction) were conducted according to Method 1311 (EPA 1990). Preliminary data indicated that 116-C-1 (Batch II) sample contained higher trace metal concentrations (arsenic, barium, chromium, and lead) than 116-C-1 (Batch I). Therefore, TCLP extraction was conducted only on Batch II sample from 116-C-1 trench. The extracts were analyzed for seven regulated metals (arsenic, barium, cadmium, chromium, lead, and selenium) by ICP-MS (PNL-ALO-280), and Hg by cold vapor atomic absorption analysis (PNL-ALO-213).

#### 4.2.7 Sequential Extraction

Sequential extractions of soils were conducted to gain some understanding of contaminant binding mechanisms with operationally defined groups of mineral forms in soils. The method (Belzile et al. 1989) consists of extracting soils sequentially with increasingly strong extractants; the fractions solubilized are characterized as "exchangeable," "carbonate-bound," "Mn-oxide bound," "Fe-oxide bound," "organic matter and sulfide bound," and "residual mineral bound." In this method, these extractants are used sequentially: 1) 1N magnesium chloride solution (pH 7) equilibrated with soil for 30 min to displace the exchangeable fraction, 2) 1M sodium acetate solution (adjusted to pH 5 with acetic acid) contacted with soil for 5 hours at room temperature to dissolve "carbonate-bound" fraction, 3) a solution of 0.1M hydroxylamine-hydrochloride and 0.1M nitric acid equilibrated with soil for 30 min at room temperature to dissolve "Mn-oxide bound" fraction, 4) a mixture of 0.04M hydroxylamine hydrochloride and 25% (v/v) acetic acid heated with soil for 6 hr at 96°C to mobilize "Fe-oxide bound" fraction, and 5) digestion of the soil for 5 hr at 85°C with 30% hydrogen peroxide (acidified to pH 2 with nitric acid) followed by a room temperature extraction with a solution of 3.2M ammonium acetate and 20% (v/v) nitric acid to release "organic-bound" or "sulfide-bound" fractions. These extraction steps were expected to provide information on specific affinities of contaminants for different types of mineral surfaces and matrices.

### 4.3 MINERALOGICAL CHARACTERIZATION

Radionuclides in soil can exist in several forms such as exchangeable, specifically adsorbed, surface-precipitated, and as part of substrate mineral structure. The ease of removal of a contaminant depends on the type of association between the contaminant and the mineralogical substrates. Sequential extraction techniques provide some information regarding the types of minerals that may exist in soils; however, these techniques do not directly identify specific minerals and their association with different contaminants.

Mineralogy of these soils was determined by using two different techniques. X-ray diffraction analyses (XRD) was used to determine the structural identity, and scanning electron microscopy with energy dispersive spectrometry (SEM-EDS) analyses was used to obtain chemical and morphological data on minerals present in the sand fraction (2 to 0.25 mm) of 116-D-1B soil. Minerals in the clay fractions ( $< 2 \mu\text{m}$ ) of 116-D-1B Batch III and 116-C-1 Batch II were identified by XRD.

The XRD of the sand fraction (2 to 0.25 mm) of 116-D-1B soil was conducted by grinding the sample into silt-sized material, packing this material into aluminum sample holders to obtain randomly oriented specimens, and scanning these samples using Cu or Co  $K\alpha$  radiation. The minerals were identified by their characteristic diffraction patterns with standard diffraction data from Joint Committee on Powder Diffraction Standards (JCPDS). The XRD of clay fractions was conducted by preparing samples oriented on glass slides. These samples consisted of clays that were K-saturated, Mg-saturated, K-saturated and heated to 550°C, and Mg-saturated and glycerated. Oriented sample specimens were scanned from 2 to 30° (2 $\theta$ ) using Co  $K\alpha$  radiation. Clay minerals were identified on the basis of the typically known changes in the oriented diffraction patterns brought about by these treatments.

The SEM-EDS analyses of minerals in the sand fractions of 116-D-1B soil were conducted on selected mineral grains of diverse morphology. First, an optical binocular microscope was used to examine mineral particles under reflected light, and mineral grains with different morphologies were picked out for SEM-EDS analyses. These mineral grains were mounted on carbon specimen stubs, and coatings of electron-conductive carbon were vacuum-deposited. Mineral grains were examined in a SEM, and images were obtained using both secondary (SE) and back-scattered electrons (BSE). The BSE images were obtained to detect different mineral inclusions in the major mineral matrix. The chemical composition of different minerals was ascertained from collecting x-ray spectra.

Autoradiography was used to identify the distribution of low-energy electron (beta particles) emitting radionuclides on edges of mica particles from 116-D-1B (Batch III) soil. The mica particles were mounted on edge and encased in polymeric resin. After the resin hardened, the surface was ground flat and coated with specially formulated photographic emulsion. The images were developed after 2 months, and were examined using a SEM.

Additionally, optical micrographs of sand fraction (2 to 0.25 mm) from 116-D-1B soil were obtained before and after attrition scrubbing and analyzed using Imageset software to determine any changes in particle morphology. This image-processing software determines the equivalent diameters of soil particles (defined as the diameter of a circle with an area equivalent to the projected area of a particle), and the roundness factor for particles (defined as the ratio of 4 times the projected area to  $\pi$  times the squared length). The roundness factor is unity for a circle and is smaller for elongated particles. The fines generated from attrition scrubbing were also analyzed by XRD to determine the identity of attritted minerals.

## 4.4 RESULTS AND DISCUSSION

### 4.4.1 Physical Characterization

The air-dry gravimetric moisture content for <2-mm-sized fractions of 116-C-1 Batch I, Batch II, and 116-D-1B Batch III soils were 2.49%, 0.85%, and 2.29%, respectively. These moisture contents are indicative of low contents of clay and organic matter in these soils.

The particle size distribution data for 116-C-1 Batch II and 116-D-1B Batch III soils are shown in Figure 4-1. The particle size distributions for both samples ranges over five orders of magnitude. However, the mass median diameters of these soils is significantly different: about 10 mm for 116-C-1 Batch II, and approximately 1.3 mm for 116-D-1B Batch III. The clay and silt contents of both these samples were very low (<6%) (Table 4-1). Even though coarse sand and gravel fractions are predominant in both samples, the Batch II material contained over twice as much (total 97.2%) coarse sand and gravel than the Batch III material. About 50% of the total mass of the Batch III material occurred in the sand fraction. Using the engineering soil classification criteria (ASTM D 2487-90), the 116-C-1 Batch II material can be classified as poorly graded gravel (group symbol: GP) and the 116-D-1B Batch III material as well-graded sand with silt (group symbol: SW-SM).

For specific gravity, the tabulated data (Table 4-2) showed that the average of the bulk soils were 2.71 and 2.79 for Batch II and Batch III respectively. These values are typical of most mineral soils. The specific gravity of sand-, silt-, and clay-size material in these soils was slightly higher (2.8 to 2.9), indicating ferromagnesian mineral-rich soils. The chemical and mineralogical analyses (data presented in later part of this section) indicated that these soils do contain Fe-bearing minerals. These specific-gravity data are typically used to assess particle settling times from suspensions.

### 4.4.2 Chemical Characterization

The TOC content of 116-C-1 Batch I and II soils was 0.113% and 0.164%, respectively (Table 4-3). The soil material from the 116-D-1B trench contained the lowest TOC (0.06%) among the three soils. These low TOC values are typical of coarse-textured soils.

Figure 4-1. Particle-Size Distribution for 116-C-1 (Batch II),  
and 116-D-1B (Batch III) Soils.

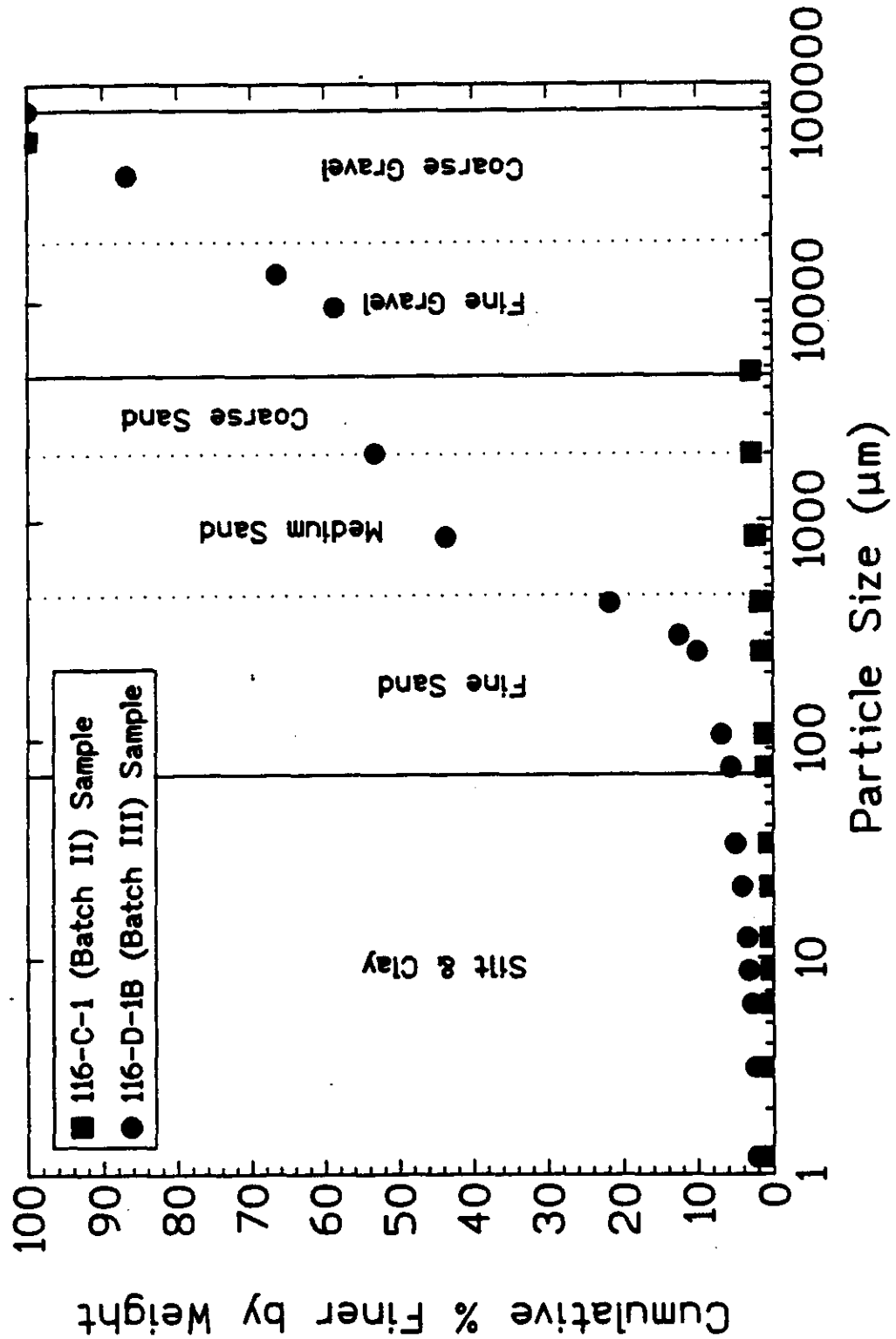




Table 4-1. Particle-Size Distribution Data for 116-C-1 Batch II and 116-D-1B Batch III Trench Soils\*.

Particle-Size Fraction**	116-C-1 Batch II	116-D-1B Batch III
	% Wt.	
Gravel (> 4.75 mm)	97.2	44.0
Coarse sand (4.75 mm - 2 mm)	0	2.8
Medium sand (2 mm - 0.425 mm)	1.1	31.4
Fine sand (0.425 mm - 0.075 mm)	0.7	16.3
Silt and clay (<0.075 mm)	1.0	5.5

\*Particle-size distribution analyses were not conducted on Batch I material.

\*\*Particle-size designations are based on standard classification method ASTM D2487-90

Table 4-2. Specific Gravity of 116-C-1 and 116-D-1B Trench Soils.

Particle-Size Fraction	116-C-1 Batch I	116-C-1 Batch II	116-D-1B Batch III
Gravel (> 4.75 mm)	--	2.70	2.66
Coarse Sand (2 mm -4.75 mm)	--	--	2.91
Medium sand, silt and clay (<2mm)	2.77	2.81	2.90
Bulk specific gravity	--	2.71	2.79

Table 4-3. Total Organic Carbon Content.

Sample	TOC (mg/kg)
116-C-1 Batch I	1130
116-C-1 Batch II	1640
116-D-1B Batch III	600

The pH values of 116-C-1 Batch I were slightly acidic, whereas the pH values of the other two samples were slightly alkaline (Table 4-4). These pH values indicated that these soils were probably fully base-saturated and that no exchangeable Al and carbonates are present.

The cation exchange capacity (CEC) measurements (Table 4-5) indicate that these soils are fully base-saturated. These data agree with the conclusions drawn from soil pH measurements. The CEC of these soils is typical of coarse-textured soils found in 100 Areas of Hanford (Benson et al. 1963). In all three soils, the dominant exchangeable cation was Ca, which accounted for 77% to 84% of the total CEC. Minor amounts of exchangeable Mg and trace quantities of exchangeable Ba, Sr, and Na were present in these soils. Together, Ca and Mg composed almost all the exchange capacity of these soils (>99%). Because of the predominance of Ca and Mg on the exchange sites, these soils are expected to flocculate during the soil-washing process.

The major and the trace element compositions of the three soils are listed in Tables 4-6 and 4-7, respectively. The major element content of these soils is typical of soils containing aluminosilicate minerals. The trace element content of these soils is within the range typically found in uncontaminated soils. Note that the total concentrations of Cr in all three soils were well below the TPL of 1600 mg/kg. Therefore, none of the trace elements in these soils is present at levels that would pose a contamination problem.

Table 4-4. pH Measurements

Sample	pH
116-C-1 Batch I	6.50
116-C-1 Batch II	7.40
116-D-1B Batch III	7.66

Table 4-5. Cation Exchangeable Capacity of 100 Area Soil Samples\*.

Sample	Exchangeable Cations (meq/100 g)					CEC meq/100 g
	Ba	Ca	Mg	Sr	Na	
116-C-1 Batch I	0.03	6.82	2.00	0.02	0.00	8.9
116-C-1 Batch II	0.02	6.65	1.65	0.02	0.09	8.4
116-D-1B Batch III	0.03	6.75	1.20	0.02	0.00	8.0

\*Measurements conducted on <2-mm material.

Table 4-6. Major Element Concentrations (%) in 100 Area Soil Samples\*.

Element	Samples		
	116-C-1 Batch I	116-C-1 Batch II	116-D-1B Batch III
Aluminum	5.70	5.11	5.67
Calcium	2.65	2.65	4.10
Iron	4.51	5.59	6.83
Potassium	1.60	1.36	1.15
Silicon	23.9	21.25	22.25
Titanium	0.64	0.65	1.02

\*Measurements of duplicate samples conducted by x-ray fluorescence spectrometry using Ag, Gd, Fe, and Zr targets.

The radionuclide data for 116-C-1 (<2-mm fractions of Batch I) soil (Table 4-8) indicated that the activities of all measured radionuclides except  $^{152}\text{Eu}$  are well below the TPL. In comparison, the <2-mm-sized fraction of soil material (2.8% of the total mass) obtained from the vicinity of the trench inlet (116-C-1, Batch II) contained activities of  $^{60}\text{Co}$ ,  $^{137}\text{Cs}$ ,  $^{152}\text{Eu}$ ,  $^{154}\text{Eu}$ , and  $^{239/240}\text{Pu}$  that exceed the TPL. Because of the small masses of contaminated <2-mm-sized fraction ( $\leq 10\%$ ), this soil fraction from the 116-C-1 (Batch I) material could be disposed following wet screening and still allow about 90% of the soil mass to be recovered. The <2-mm-sized fraction from the 116-D-1B trench, however, constituted a significant part of the total mass (53.1%); therefore, soil from this trench may need soil-washing to reduce the activity of  $^{60}\text{Co}$ ,  $^{137}\text{Cs}$ , and  $^{152}\text{Eu}$  to meet the TPL.

These data indicated that the <2-mm-size fractions of these soils had in common,  $^{60}\text{Co}$ ,  $^{137}\text{Cs}$ , and  $^{152}\text{Eu}$  as the contaminants. Therefore, the >2-mm fractions of these soils were analyzed for these three ( $^{60}\text{Co}$ ,  $^{137}\text{Cs}$ , and  $^{152}\text{Eu}$ ) radionuclides.

The mass concentrations of the contaminant radionuclides in these soils were calculated from the specific-activity data (Table 4-9). These mass concentrations are in the parts per trillion range; therefore, these contaminants are probably present in various adsorbed or coprecipitated forms and not as distinct pure solid phases.

The TCLP extraction data (Table 4-10) showed that the extract concentrations of all eight regulated elements were orders of magnitude lower than the regulatory limit. These data confirmed the conclusion drawn from the total trace element analyses that none of the trace elements is present at concentration levels to be considered a contaminant. Therefore, additional TCLP tests were not conducted on treated soil fractions. Radionuclides,  $^{60}\text{Co}$ ,  $^{137}\text{Cs}$ , and  $^{152}\text{Eu}$  and  $^{154}\text{Eu}$  present in 116-D-1B (Batch III) soil (<0.25-mm size fraction) were leachable at very low activity levels. Also, the trace level leachability indicated that in these soils chromium is present in its reduced form (Cr III).

Table 4-7. Trace Element Concentrations (mg/kg) in 100 Area Soil Samples\*.

Trace Element	Samples			Range of Concentrations in Uncontaminated Soils*
	116-C-1 Batch I	116-C-1 Batch II	116-D-1B Batch III	
Antimony	<16	<19	<19	0.2 - 10
Arsenic	4	7	<2	0.1 - 40
Barium	729	753	632	100 - 3000
Cadmium	<12	<13	<14	0.01 - 2
Chromium (Total)	56	236	58	5 - 1500
Copper	44	50	61	2 - 250
Lead	13	101	13	2 - 300
Manganese	847	1114	1154	20 - 10000
Nickel	26	37	24	2 - 750
Rubidium	63	61	43	20 - 1000
Selenium	<1	<1	<1	0.01 - 12
Silver	<10	<12	<12	0.01 - 8
Strontium	401	415	377	4 - 2000
Uranium	5	<5	9	0.7 - 9
Vanadium	165	161	295	3 - 500
Zinc	88	855	138	1 - 900
Zirconium	211	209	205	60 - 2000

\*Measurements of duplicate samples conducted by x-ray fluorescence spectrometry using Ag, Gd, Fe, and Zr targets.

\*\*Bowen (1979).

Table 4-8. Radionuclide Data for 100 Area Soil Samples\*.

Radionuclide (pCi/g)	116-C-1 Batch I	116-C-1 Batch II	116-D-1B Batch III	Accessible Soil Activity Limits**
<sup>60</sup> Co	7	525	15	7.1
<sup>134</sup> Cs	<0.8	<10	<2	10
<sup>137</sup> Cs	0.74	5495	205	30
<sup>152</sup> Eu	28	2320	177	15
<sup>154</sup> Eu	4.4	337	17	14
<sup>155</sup> Eu	0.54	70	1.4	630
<sup>90</sup> Sr	<0.2	115	12.5	2800
<sup>235</sup> U	0.06	0.06	0.11	170
<sup>238</sup> U	1.31	1.23	2.38	370
<sup>239/240</sup> Pu	0.08	414	2.74	190

\*Analyses conducted on &lt;2-mm-size material.

\*\*Table 6-2, WHC(1988).

Table 4-9. Activities and Concentrations of Contaminant Radionuclides in 116-C-1 and 116-D-1B Trench Soils\*.

Radio-nuclide	116-C-1 Batch I		116-C-1 Batch II		116-D-1B Batch III	
	Activity	Concentration	Activity	Concentration	Activity	Concentration
	pCi/g	mg/kg	pCi/g	mg/kg	pCi/g	mg/kg
<sup>60</sup> Co	7	6.17x10 <sup>-9</sup>	525	4.63x10 <sup>-7</sup>	15	1.32x10 <sup>-8</sup>
<sup>137</sup> Cs	0.74	8.56x10 <sup>-9</sup>	5495	6.36x10 <sup>-5</sup>	205	2.37x10 <sup>-6</sup>
<sup>152</sup> Eu	28	1.55x10 <sup>-7</sup>	2320	1.28x10 <sup>-5</sup>	177	9.77x10 <sup>-7</sup>
<sup>154</sup> Eu	4.4	1.60x10 <sup>-8</sup>	337	1.23x10 <sup>-6</sup>	17	6.46x10 <sup>-7</sup>
<sup>239/240</sup> Pu	0.08	1.29x10 <sup>-6</sup>	414	6.67x10 <sup>-3</sup>	2.74	4.41x10 <sup>-5</sup>

\*Analyses conducted on &lt;2-mm material.

Table 4-10. Analyses of Extracts from Toxicity Characteristics Leaching Procedure.

Element	Soil Sample			EPA Regulatory Level (mg/L)
	116-C-1 Batch II* (mg/l)	116-D-1B Batch III* (mg/l)	116-D-1B Batch III** (mg/l)	
Arsenic	0.20	0.20	ND	5.0
Barium	0.35	0.29	0.47	100.0
Cadmium	0.01	0.02	0.01	1.0
Chromium	<0.02	<0.02	0.04	5.0
Mercury	<0.0004	<0.0004	ND	0.2
Silver	0.03	<0.01	0.01	1.0
Lead	<0.06	<0.06	0.03	5.0
Selenium	<0.2	<0.2	ND	1.0

\*Extractions conducted on <2-mm-sized soil fractions.

\*\*Extraction conducted on <0.25-mm-sized soil fraction. The activities of  $^{60}\text{Co}$ ,  $^{137}\text{Cs}$ ,  $^{152}\text{Eu}$ , and  $^{154}\text{Eu}$  in the leachate were <56, 37, 200, and <90 pCi/L respectively.

ND: Not determined

The results of different extractive treatments (Table 4-11) showed that the first extractive step removed minor fractions of the radionuclide activity from both soils. The fraction of  $^{137}\text{Cs}$  displaced in this step was a mere 0.5% from the higher activity 116-C-1 Batch II soil and 2% from the lower activity 116-D-1B Batch III soil. Similarly, 4% to 5% of  $^{152}\text{Eu}$  and 2% to 16% of  $^{60}\text{Co}$  were mobilized by the salt solution.

The fourth extraction step involving heating and digestion with a reductive acid solution released more  $^{137}\text{Cs}$  from both soils than the combined total amount obtained from the three previous extractive steps. The fractional activities of  $^{60}\text{Co}$  removed in this extractive step were similar to the amounts extracted in the previous reductive extractive step. The extractability of  $^{152}\text{Eu}$  from the two soils differed significantly. A major fraction (about 79%) of the total burden of this radionuclide was removed from the 116-D-1B Batch III soil as compared to about 26% removed from the 116-C-1 Batch I material.

The final two-stage oxidative-acid digestion step released more  $^{137}\text{Cs}$  from both soils than any of the four previous extractions. Removal of  $^{60}\text{Co}$  from these soils in this step matched the amounts that were released in the previous step. Minimal and below detectable amounts of  $^{152}\text{Eu}$  were mobilized from 116-C-1 Batch II, and 116-D-1B Batch III soils, respectively.

The fractional activities of all three radionuclides released during the second step (acid-sodium acetate solution) were similar to the fractional activities mobilized during the initial step. Distinct differences in extractable fractions of radionuclides were observed following the third extractive step (reductive acid soluble treatment). In this step,  $^{137}\text{Cs}$  was mobilized in minor quantities that were similar to the amounts released during the two previous steps, whereas noticeable amounts (15% to 17%) of  $^{60}\text{Co}$  were extracted from the soils. Also, this extractive step showed significant differences in the amounts of  $^{152}\text{Eu}$  released from the two soils. About 26% of total  $^{152}\text{Eu}$  in 116-C-1 Batch I was released by this extraction; however, no detectable quantities of this radionuclide were mobilized from the 116-D-1B Batch III soil.

The residual activities of the three radionuclides in these soils showed all the sequential extractive steps in total removed significant fractions of  $^{152}\text{Eu}$  (61% to 92%) and  $^{60}\text{Co}$  (57% to 76%) but mobilized only 22% to 40% of  $^{137}\text{Cs}$  activity. These differences in total extractability indicated that in these two soils, significant fractions of  $^{137}\text{Cs}$  are associated with recalcitrant solid phases.

These sequential extraction data show that only minor fractions of these radionuclides are tied to "exchangeable" or "carbonate-bound" phases. About equal amounts of  $^{60}\text{Co}$  in both the soils were bound with "Mn-oxide," "Fe-oxide," and "oxidizable acid-soluble solid" phases. Extractable fractions of  $^{137}\text{Cs}$  in these trench soils appeared to be associated primarily with "Fe-oxide" and "oxidizable acid-soluble solid" fractions. In 116-C-1 Batch II soil, extractable  $^{152}\text{Eu}$  appeared to be bound in about equal proportion with "Mn-oxide" and "Fe-oxide" phases. By contrast, the major extractable fraction of  $^{152}\text{Eu}$  in the 116-D-B Batch III trench soil appeared to be bound mainly with the "Fe-oxide" phase.

#### 4.4.3 Mineralogical Characterization

Plagioclase feldspar and quartz were the major minerals (30% to 70% by mass) found in the 2- to 0.25-mm-sized fraction and thus constitute the principal mineral matrix of the 116-D-1B soil (Table 4-12). Micaceous minerals (biotite, illite, and muscovite), kaolinite, hornblende, and Fe-titanate were present in minor quantities (3% to 10% by mass). Minerals present in trace quantities (<3%) were smectite, vermiculite, chlorite, Fe-oxide, potassium feldspar, and sodium feldspar. SEM-EDS data showed that sand-size mineral particles in general contained other minerals as inclusions. For instance, quartz, hornblende, and mica were found both as distinct particles and as inclusions in plagioclase feldspar grains (Figure 4-2). Similarly, K-feldspar also existed as separate particles and as inclusions in Na-feldspar matrix (Figure 4-3). Fe-titanate was found exclusively as inclusions in plagioclase feldspar (Figure 4-4). Optical microscopy showed that many mineral grains had extensive coatings of white material and reddish-yellow stains (Figure 4-5). Using stereo pairs of electron micrographs, the thickness of the white coatings was estimated to range from 0.02 to 0.18 mm. Energy-dispersive x-ray analyses indicated that the white coatings consisted of an aluminosilicate mineral, kaolinite (Figure 4-6), and that the reddish-yellow

material was Fe-oxide. The trace phase mineralogy (clay fraction) of both soils (116-C-1 Batch II and 116-D-1B Batch III) was similar in that they both contained smectite, vermiculite, chlorite, kaolinite, plagioclase feldspar, and quartz.

The types of associations that are known to occur between the contaminant radionuclides ( $^{60}\text{Co}$ ,  $^{137}\text{Cs}$ , and  $^{152}\text{Eu}$ ) and the minerals that were identified in these soils are listed in Table 4-13. The exchangeable sites on these minerals typically consist of ionizable surface hydroxyls that adsorb ions. Increasing pH results in more negatively ionized exchange sites, thus resulting in increased cation exchange capacities of minerals. Additionally, smectites possess a number of interlayer exchange sites that are not affected by the pH. These sites originate as a result of isomorphous substitutions within the tetrahedral and octahedral cationic positions within the smectite structure. Depending on the strength of binding, fractions of cations occupying these exchange sites can easily be displaced by electrolytes. The sequential extraction data (see Table 4-11) showed that the exchangeable fractions of radionuclides in these soils constituted only a minor fraction of the total activity.

Extensive studies have shown that wedge sites on micas and vermiculite that have uniquely high affinities for Cs (Scott and Smith 1987). For instance, based on adsorption experiments on various clay minerals, Sawhney (1964) concluded that Cs is preferentially "fixed" into the wedge sites of micas and vermiculites and this fixed Cs was not exchangeable. Therefore, mobilization of Cs occupying these wedge sites can only be accomplished by disrupting and/or dissolving the mineral structures. The autoradiograph (Figure 4-7) showed that the edges of mica (possibly wedge sites) contained concentrations of radionuclides. Because these wedge-sites have very high affinities for cesium, this autoradiograph might represent the  $^{137}\text{Cs}$  distribution on these sites. Sequential extraction data (see Table 4-11) confirmed that more Cs (20% to 37%) can be extracted by treating soils with hot reductive, and oxidative acids. However, the standard extractants used in this procedure appeared incapable of releasing substantial fractions (60% to 80%) of Cs that were associated with wedge and structural sites of minerals.

By contrast, the final three extraction steps mobilized substantial fractions of Eu (75% to 80%) and significant fractions of Co (53% to 56%) indicating that these radionuclides were mobilized from adsorbed and structural sites of minerals which were susceptible to the specific acid extractive treatments used in this sequential extraction procedure.

The data from radionuclide analyses (see Table 4-9), the sequential extraction (see Table 4-11), and mineralogical analyses (Table 4-12) show that the contaminants ( $^{60}\text{Co}$ ,  $^{137}\text{Cs}$ , and  $^{152}\text{Eu}$ ) are present in trace concentrations and that these radionuclides appear to be associated with varying affinities on different mineral sites. These characterization data were used to formulate potential soil-washing treatments to effectively release the contaminant radionuclides from selected soil size fractions.



Table 4-11. Sequential Extraction Data for 116-C-1 and 116-C-1B Trench Soils.

Extraction Step	116-C-1 Sample Batch II			116-D-1B Sample Batch III		
	<sup>60</sup> Co	<sup>137</sup> Cs	<sup>152</sup> Eu	<sup>60</sup> Co	<sup>137</sup> Cs	<sup>152</sup> Eu
I. MgCl <sub>2</sub> extractable %	2.1	0.5	4.3	15.5	2.0	5.1
II. Acidic sodium acetate soluble %	2.3	0.8	2.2	5.4	2.4	7.3
III. Reductive-acid soluble %	17.1	1.9	26.4	15.2	3.9	--
IV. Reductive-acid soluble (heated) %	19.6	7.8	26.4	20.6	11.7	79.3
V. Oxidative-acid soluble (heated) %	16.1	11.3	2.2	19.8	21.1	--
Residual %	42.8	77.7	38.5	23.5	58.9	8.3
Initial Activity (pCi/g)	428	4790	2270	17	180	170

Table 4-12. Mineralogy of 116-D-1B Trench Soil

Mineral Phases		
Major Phases	Minor Phases	Trace Phases
Plagioclase Feldspar	Mica (biotite, illite, muscovite)	K-Feldspar
Quartz	Kaolinite	Na-Feldspar
	Hornblende	Smectite
	Fe-titanate	Vermiculite
		Chlorite
		Fe-oxide

Table 4-13. Types of Contaminant-Mineral Associations.

Contaminant	Type of Sites in Minerals		
	Exchangeable Site	"Wedge" Site	Structural Site
Cs	Smectite, chlorite, kaolinite, Fe-oxide	Micas, vermiculite	K-feldspars, micas
Eu	Smectite, chlorite, kaolinite, micas, vermiculite, Fe-oxide	--	Feldspars
Co	Smectite, chlorite, kaolinite, micas, vermiculite	--	Smectite, chlorite, micas, vermiculite, Fe-oxide

Figure 4-2. A Scanning Electron Micrograph (Back-Scattered Mode) of a Plagioclase Feldspar Particle. The bright inclusions are hornblende, and the darker areas are quartz inclusions.

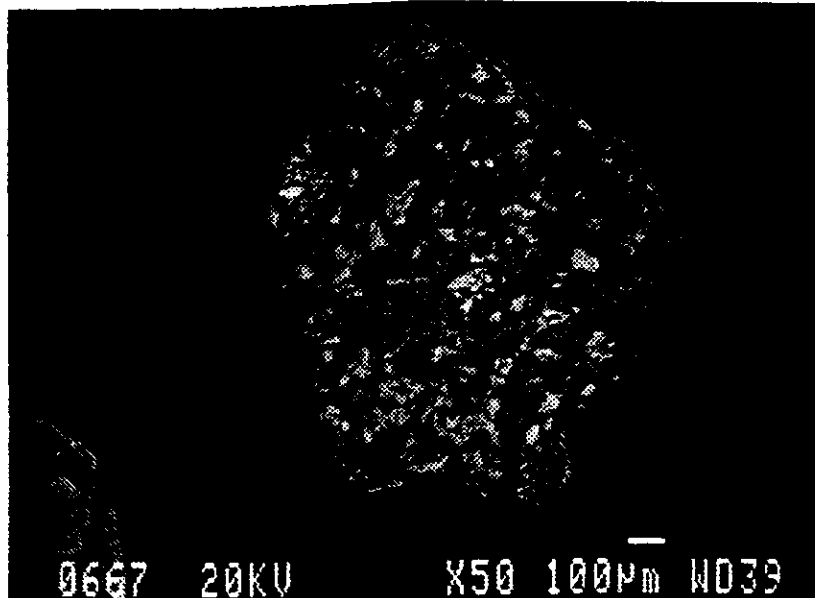


Figure 4-3. A Scanning Electron Micrograph (Back-Scattered Mode) of a Sodium Feldspar Fragment with Potassium Feldspar Inclusions (Lighter Areas).

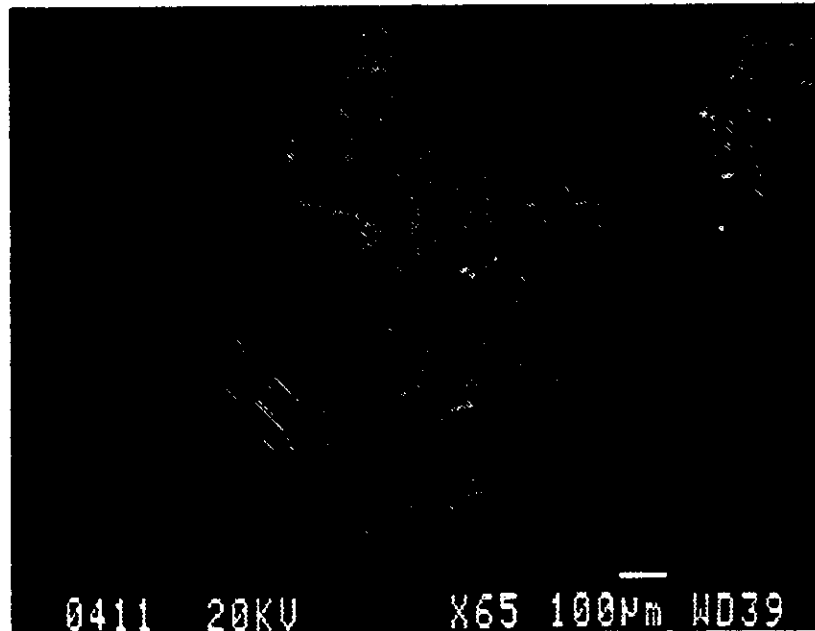


Figure 4-4. A Scanning Electron Micrograph of a Plagioclase Feldspar Grain with Inclusions of Hornblende (Light Areas), and Fe-Titanates (Very Bright Regions).

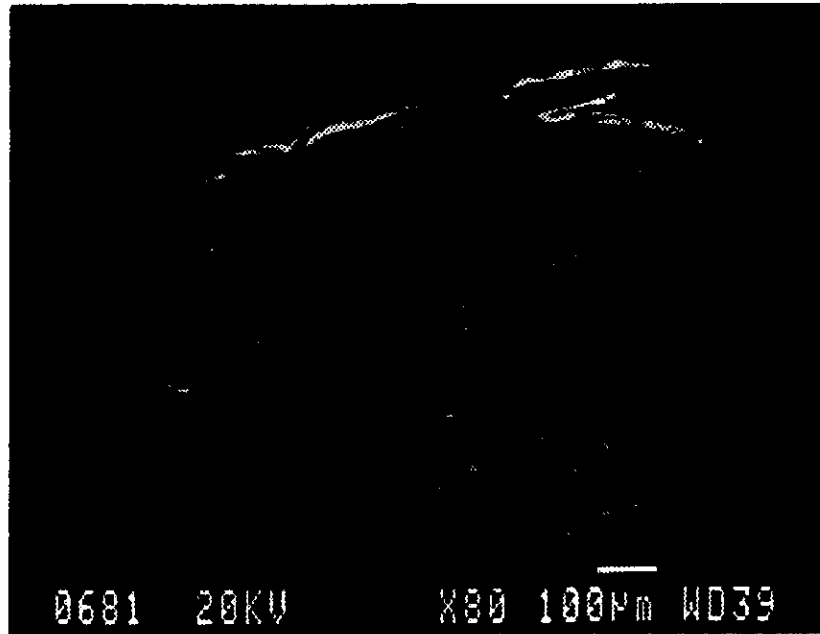
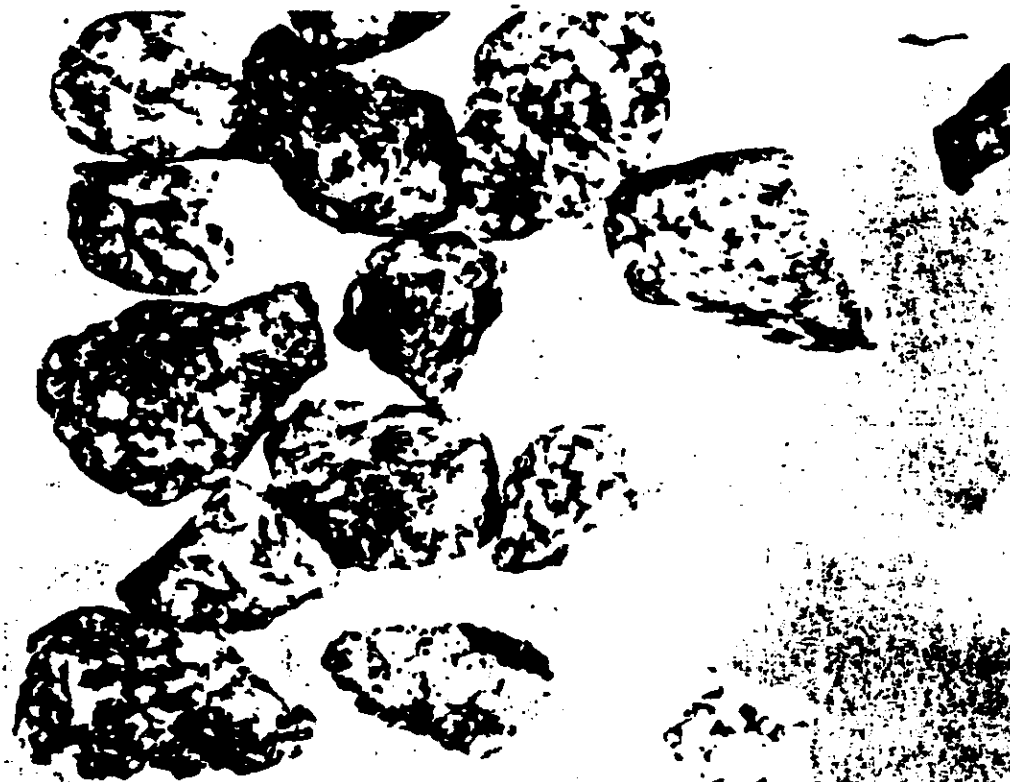


Figure 4-5. Optical Micrograph of Soil Particles With White Coatings and Reddish-Yellow Stains.



9413221.0048

Figure 4-6. A Scanning Electron Micrographic of Kaolinite Coatings Removed from the Sand-Sized Particles from 116-D-1B Soil.

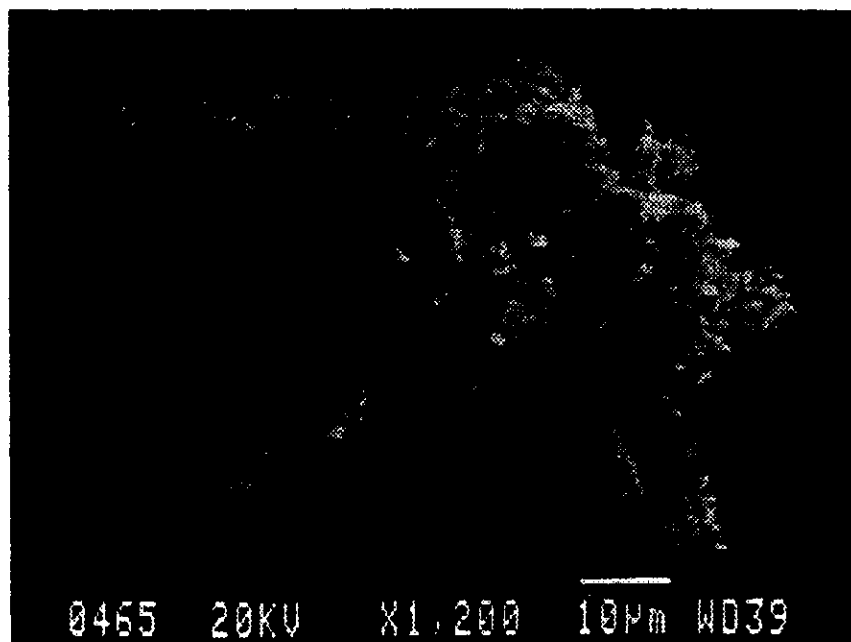
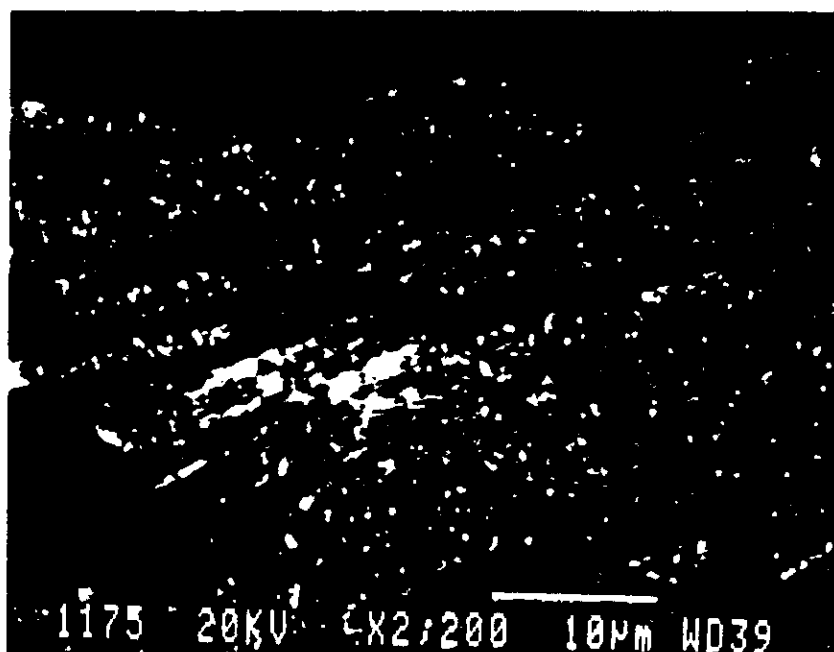


Figure 4-7. Autoradiograph of Radionuclides located on Mica Edge.



## 5.0 WET-SCREENING

### 5.1 OBJECTIVE

The goal of this test was to find the extent to which contaminants are associated with various particle size fractions of soils. In soils, the finer-sized fractions, because of their larger surface areas per unit mass, usually contain larger fractions of contaminants. By preferential removal of fine fractions through wet-sieving (i.e., soil washing), significant fractions of the total soil contamination can be isolated for disposal. The wet-screening test was conducted to determine the mass distribution of contaminants within various size fractions of soil samples, within Batch I and Batch III, and from 116-C-1 and 116-D-1B, respectively. Wet-sieving on <2-mm-sized material from Batch II samples (trench 116-C-1) was not conducted because this size fraction constituted only 3% of the total mass and contained very elevated radionuclide activities. Also, radionuclide activities were measured in samples of water-washed gravel fractions from all three soil samples. The data generated were necessary to assess the contaminant mass in each of the soil size fractions and the activity released into wash water during the sieving process, a proxy for physical soil washing. These data were used to identify the soil fractions that needed additional treatment such as attrition scrubbing and/or chemical extraction to reduce the contaminant activities to meet the TPL.

### 5.2 EQUIPMENT AND PROCEDURES

All wet-screening experiments were conducted with a Gilson Wet-Vac unit. The unit consists of a sieve-nesting cylinder, filter holder, and a discharge cone assembly mounted on a vibrating frame (Figure 5-1). This unit also includes a water spray system that can be operated either in automated or manual mode. Additionally, vacuum can be applied to the sieve assembly to hasten the screening process. The Gilson wet-sieving system can be operated either by recirculating the wash water or by once-through use of water that drains into an external tank. When wet-sieving was conducted in the water recirculating mode, it was necessary to use a filter to screen out the <25- $\mu$ m particles from wash water to prevent clogging the water recycling system. Wet-sieving operations using this unit can be conducted at different vibration intensities to optimally agitate the wet soil particles.

The procedure used for wet-screening was similar to the ASTM Method D 422-63 except for the following modifications. Because the objective of this test was to examine the contaminant distribution among particle fractions, no dispersant was used. The suggested use of a mixture of sodium hexametaphosphate and sodium hydroxide was omitted because dispersants tend to release and redistribute the contaminants between soil and aqueous phases.

Figure 5-1. Gilson Wet-Vac Wet-Sieving Unit.



All wet-sieving experiments were conducted with a set of sieves [9.5 mm (3/8 in.), 2 mm (No. 10), 0.25 mm (No. 60), and 0.075 mm (No. 200)]. Initial experiments were conducted in the wash water recycling mode; therefore, the filter section was used to retain the 20 to 25  $\mu\text{m}$  fines passing the No. 200 sieve. At the end of the sieving cycle (which lasted approximately 40 to 80 min, soil fractions retained on each sieve were rinsed with fresh deionized distilled water until the wash water was clear. The soil fractions retained on the sieves were oven-dried at  $105 \pm 5^\circ\text{C}$  and weighed. The oven-dried soil fractions were composited to represent >2-mm, 2.00- to 0.25-mm, and 0.25- to 0.075-mm-sized fractions. Aliquots of soil fractions <20 to 25  $\mu\text{m}$  were filtered out of wash water and dried. The quantity of soil that could be optimally sieved in each cycle depended on the texture of the soil and ranged from 75 to 150 g. Exceeding the optimal sample weight for a soil caused sieve blinding. Therefore, wet-sieving was conducted a number of times to accumulate sufficient quantities of soil in each sieve fraction. These soil fractions, and the wash water were analyzed for the contaminants of interest. Mass balances were computed for weight fractions and the activity balances were computed from the radionuclide data.

The preliminary data indicated that wet sieving in the wash water recycling mode required 8 to 9 L of water to process approximately 0.5 kg of soil, and, because of filter section blinding each sieving cycle extended from 40 to 80 min. Additionally, the radionuclide counting data of sieved soil fractions suggested that the filter paper may have adsorbed part of the total activity. Therefore, all subsequent wet sieving was conducted in once-through mode without the filter section. The once-through-mode sieving required only 3 to 4 L of water during 40 min to completely wet-sieve about 0.5 kg of soil.

The radionuclide activity data on the gravel fractions of all soils were obtained by manually washing several kilogram quantities of samples and gamma counting the oven-dried material.

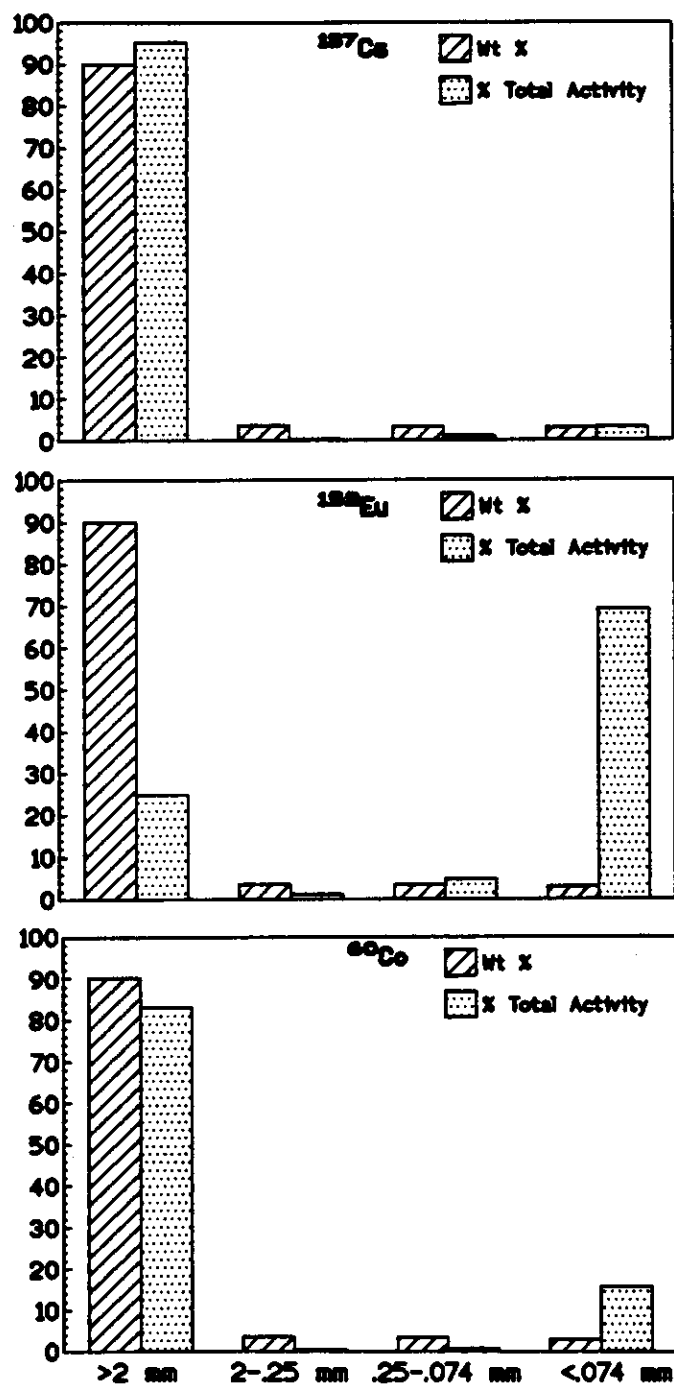
### 5.3 RESULTS AND DISCUSSION

The wet-sieving data for 116-C-1 Batch I (Table 5-1) indicated that all size fractions except the finest ( $<0.074$  mm) contained radionuclide activities that were significantly below the TPL. In this soil,  $>2$ -mm size fraction (90% of the mass) contained major fractions of total activities of  $^{60}\text{Co}$  (83 %) and  $^{137}\text{Cs}$  (95 %) (Figure 5-2). The bulk of the  $^{152}\text{Eu}$  activity (69 %) in this soil however, was present in the finest size fraction ( $<0.07$ -mm). The average radionuclide activities of the bulk soil were also below the TPL. Therefore, soil washing is not necessary if the bulk of the soil in this trench is similar to Batch I material (sampled near the middle of the trench).

The gravel fraction ( $>2$  mm) from 116-C-1 Batch II soil (constituting about 97% of the total soil mass) contained  $^{60}\text{Co}$ ,  $^{137}\text{Cs}$ , and  $^{152}\text{Eu}$  activities of 17, 726, and 46 pCi/g, respectively. These higher activities are well above the TPL values of 7.1, 30, and 15 pCi/g, respectively, for these radionuclides. These enhanced activities may be typical of soils near the trench inlet and may constitute a minor mass of the whole 116-C-1 trench. Additional soil-washing treatments such as autogenous grinding and/or chemical extraction may be necessary for reducing the radionuclide activities in the gravel fraction.

The radionuclide distribution in 116-D-1B (Batch III) soil showed typical particle-size dependence (see Table 5-1), namely increasing activities with decreasing particle size. In this soil,  $<0.25$ -mm-sized particle fractions contained 68 %, 53 %, and 75 % of the total  $^{60}\text{Co}$ ,  $^{137}\text{Cs}$ , and  $^{152}\text{Eu}$  activities in the whole soil (Figure 5-3). Separating the  $<0.25$ -mm-sized fraction (only 10.8 % of the soil mass) will, therefore remove significant fractions of these radionuclides from the soil. The composite of  $>2$ -mm-sized and 2- to 0.25-mm-sized fractions (89.2 % of mass) contains  $^{60}\text{Co}$ ,  $^{137}\text{Cs}$ , and  $^{152}\text{Eu}$  activities of 2, 56, and 24 pCi/g, respectively. The Co activity in the composite is below the TPL; however, the activities of Cs and Eu need to be reduced for the composite to meet the TPL for these two contaminants. The sand fraction (2 to 0.25 mm) contains a higher fraction of the activities than the gravel fraction ( $>2$  mm); therefore, the sand fraction needs additional treatment to bring the overall

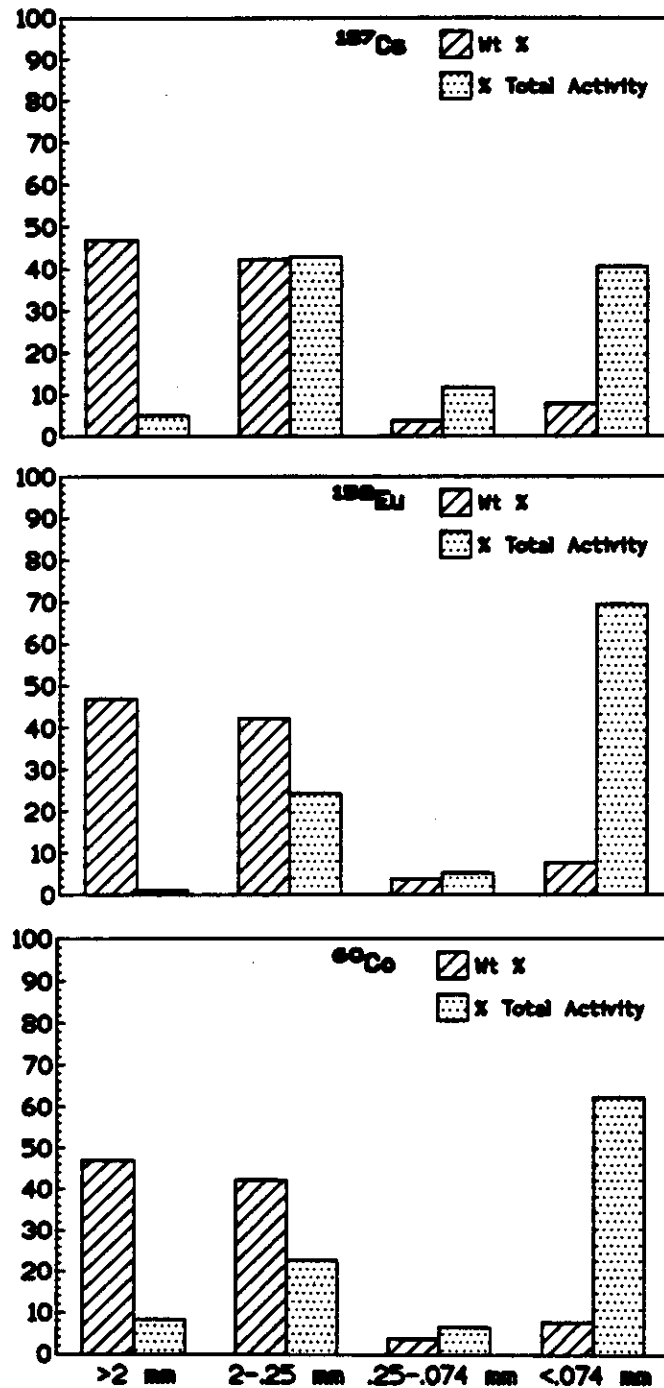
Figure 5-2. Mass and Radionuclide Distribution in 116-C-1 (Batch I) Soil.



9443221-0053



Figure 5-3. Mass and Radionuclide Distribution in 116-D-1B (Batch III) Soil.



9443221.0054

Table 5-1. Contaminant Radionuclide and Mass Distribution Data for 116-C-1 Batch I and 116-D-1B Batch III Soils\*.

Particle Size	116-C-1 Batch I				116-D-1-B Batch III			
	<sup>137</sup> Cs	<sup>152</sup> Eu	<sup>60</sup> Co	Wt %	<sup>137</sup> Cs	<sup>152</sup> Eu	<sup>60</sup> Co	Wt %
	pCi/g				pCi/g			
> 2 mm	2	0.6	3.2	90.0	11	2	<1	46.9
2 mm - 0.25 mm	0.2	0.8	0.5	3.6	105	48	3	42.3
0.25 mm - 0.074 mm	0.6	3	0.8	3.4	325	117	10	3.7
<0.074 mm	2	50	18	3.0	590	819	49	7.1
Bulk Soil	1.9	2.2	3.5	100.0	103.5	83.7	5.6	100.0

\*The gravel fractions (>2-mm-sized fraction) were water-washed manually.

activities in the composite to below TPL values. Calculations showed (Figure 5-4) that Cs and Eu activities in 2- to 0.25-mm-sized fraction need to be reduced by about 50% and 40%, respectively for the composite to meet the TPL.

The wet-sieving experiments provided critical data to assess the necessary soil-washing treatments for each soil. For 116-C-1 Batch I soil, wet-sieving alone is sufficient to recover about 97% of the soil that meets the TPL requirements. The 116-C-1 Batch II soil has high radionuclide activities in both coarse and fine fractions; therefore, the gravel fraction (97.2% of the total mass) needs additional treatment to meet the TPL. The finer fractions (<0.25 mm) from trench 116-D-1B contain higher activities. These fractions (10.8% of the total mass) can be fractionated for disposal. Additional treatments of the 2- to 0.25-mm fraction (42.3% of the total soil mass) using attrition scrubbing or chemical extraction are necessary for the composite fraction of this soil (>2-mm and the 2- to 0.25-mm-sized fractions) to achieve the TPL for the contaminant radionuclides.

## 6.0 ATTRITION SCRUBBING

### 6.1 OBJECTIVE

The attrition scrubbing tests were conducted to examine whether contaminants can be removed from the surfaces of soil particles through scrubbing action. These tests were not necessarily designed to provide data to directly design full-scale equipment. However, the results should show whether some form of attrition scrubbing is beneficial in partitioning contaminants to the fine fraction. We evaluated a number of parameters that affect attrition scrubbing, including solids density, impeller speed, residence time, and the use of electrolytes. Attrition scrubbing tests are usually conducted on sand-sized material. All the scrubbing tests were conducted on 2- to 0.25-mm-sized fractions (about 40% of the total mass) from 116-D-1B trench soil.

### 6.2 EQUIPMENT AND PROCEDURES

The attrition scrubbing tests were performed in a laboratory-scale attrition scrubber fabricated from a high-torque, servo-controlled, stir motor with a stainless steel shaft and two three-bladed 7.5-cm-diameter impellers (Figures 6-1 and 6-2). The impellers were fixed on the end of the steel shaft with the blades aligned with opposing pitch. This configuration maximizes the particle-to-particle contact and results in the desired scrubbing action. The motor speed can be continuously adjusted from 20 to 900 RPM, and the speed is maintained by a servo-control loop to ensure reproducibility between tests. The motor controller also has a built-in timer to allow the contact times to be controlled precisely. The scrubber can be programmed to run at either a fixed RPM or specified torque. The mix containers used in these experiments were 1-L rectangular containers.

All scrubbing tests were conducted on approximately 500-g loads of previously wet-sieved and air-dried 2- to 0.25-mm-sized fractions of 116-D-1B trench soil. Preliminary observations indicated that particles were moving effectively when scrubbing was conducted with impeller speed set at 900 RPM. The first set of experiments was conducted to identify the range of pulp densities for optimum scrubbing to occur. These tests were conducted at 900 RPM for 15 min with deionized distilled water added to achieve pulp densities ranging from 75% to 85% (weight-percent solids). Based on the data from these tests, subsequent tests were conducted for variable residence times at fixed optimum pulp density and impeller speed. During these tests, torque values were recorded at 5-min intervals to track the total energy input for each experiment. The energy input per unit mass (HP-min/lb) was computed by the relationship (Shigley and Mischke 1989):

$$\text{HP-min/lb} = (T \times n \times t) / (1.008 \times 10^6 \times M) \quad (6-1)$$

Where T is the torque in units of oz.in, n is the shaft speed in revolutions per min (RPM), t is the scrubbing time in min, and M is the mass of soil being scrubbed.

Figure 6-1. Laboratory-Scale Attrition Scrubber with Servo-Controlled Motor.

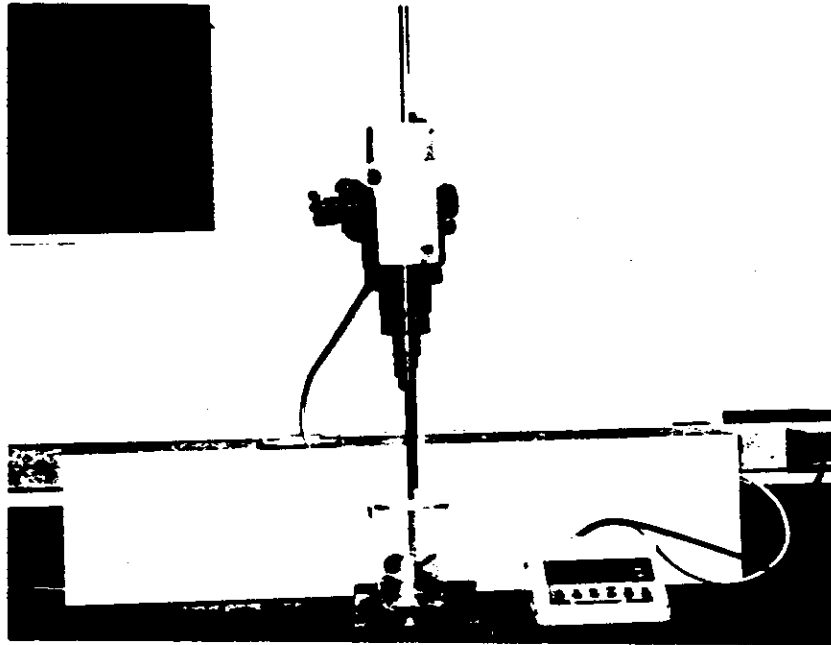
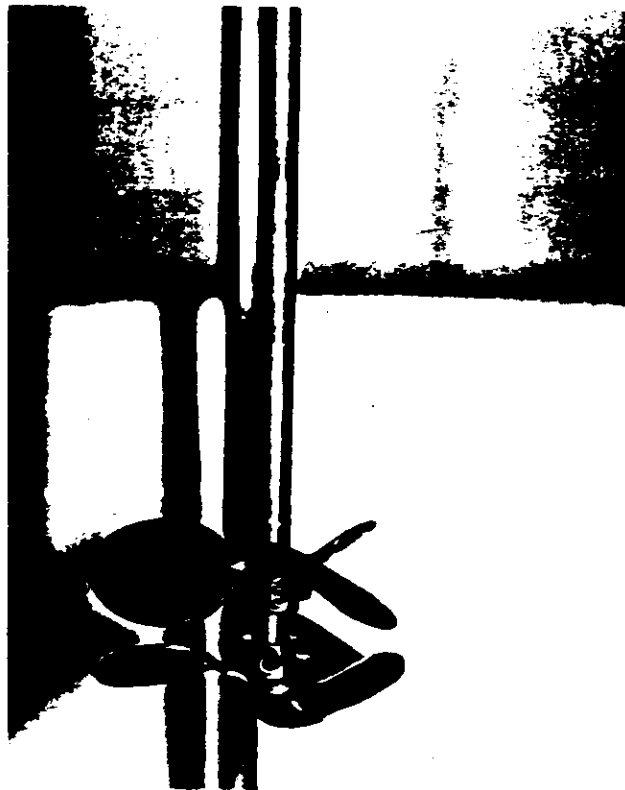


Figure 6-2. Close-up View of Opposing Pitch Impellers.



Additional tests were also conducted in electrolyte medium (0.5M ammonium citrate with citric acid) to compare the scrubbing performance obtained with deionized water.

All scrubbed soil samples were wet-sieved through a 0.25-mm sieve and both scrubbed (2- to 0.25-mm) and fine (<0.25-mm) sized fractions were oven-dried at  $105 \pm 5^\circ\text{C}$ . Representative subsamples were then drawn for radionuclide ( $^{60}\text{Co}$ ,  $^{137}\text{Cs}$ , and  $^{152}\text{Eu}$ ) measurements. Wash water was filtered through a 0.45- $\mu\text{m}$  filter, and aliquots of water were also counted for radionuclide activity.

### 6.3 RESULTS AND DISCUSSION

Preliminary data (Table 6-1) showed that the activities of  $^{60}\text{Co}$ ,  $^{137}\text{Cs}$ , and  $^{152}\text{Eu}$  in the 2- to 0.25-mm-sized soil fraction could be reduced by attrition scrubbing. Significant reduction (>70%) in  $^{60}\text{Co}$  activity was achieved in all tests; however, the reduction in activities of  $^{137}\text{Cs}$  and  $^{152}\text{Eu}$  appeared to be related to pulp density (Figure 6-3). The data indicated that the reduction in activity of these two radionuclides increased with increasing pulp densities, with optimum reduction (an average of 22% and 50% for  $^{137}\text{Cs}$  and  $^{152}\text{Eu}$ , respectively) achieved when scrubbing was conducted at a pulp density of about 83%.

Additional data from experiments conducted for identifying the optimum energy inputs (and residence times) at a fixed pulp density of 83% are listed in Table 6-2. The data showed that  $^{60}\text{Co}$ ,  $^{137}\text{Cs}$ , and  $^{152}\text{Eu}$  activities in the 2- to 0.25-mm-sized soil fraction decreased as a function of increasing input of energy. The reduction in  $^{60}\text{Co}$ ,  $^{137}\text{Cs}$ , and  $^{152}\text{Eu}$  activities (about >90%, 26%, and 61%, respectively) became asymptotic at energy inputs exceeding about 1.43 HP-min/lb (Figure 6-4). The energy expended (1.43 HP-min/lb) also generated heat and at the end of experiments, the scrubbed solids had attained temperatures ranging from 40 to  $55^\circ\text{C}$ . The quantities of fines (<0.25 mm) generated ranged from 4% to 11% by weight and also appeared to reach a plateau beyond the same input of scrubbing energy (Figure 6-5). These data suggested that attrition scrubbing becomes less efficient as increasing amounts of fines are generated. The decreasing scrubbing efficiency with increasing residence time also corresponds to continually decreasing torque values (Figure 6-6). The torque data suggested that when the amount of fines generated reached a critical value (about 9% for 116-D-1B soil), the attritive action between coarse particles was reduced to the extent that no further reductions in radionuclide content were noted.

Optical microscopic examination of the attrition-scrubbed coarse material showed (Figure 6-7) significant reduction in the whitish coatings that were observed on initial material (Figure 4-5). Therefore, a significant part of radionuclide activities appeared to be associated with these coatings. The SEM-EDS indicated that these whitish coatings are aluminosilicates (see Figure 4-5). Based on these data and x-ray diffraction analyses, these coatings were identified as kaolinite, a clay mineral. The SEM-EDS also showed that the soil particles also contained micaceous minerals (Figure 6-8). Considerable data (Scott and Smith 1987) indicate that micaceous minerals have very strong adsorption affinity for Cs.

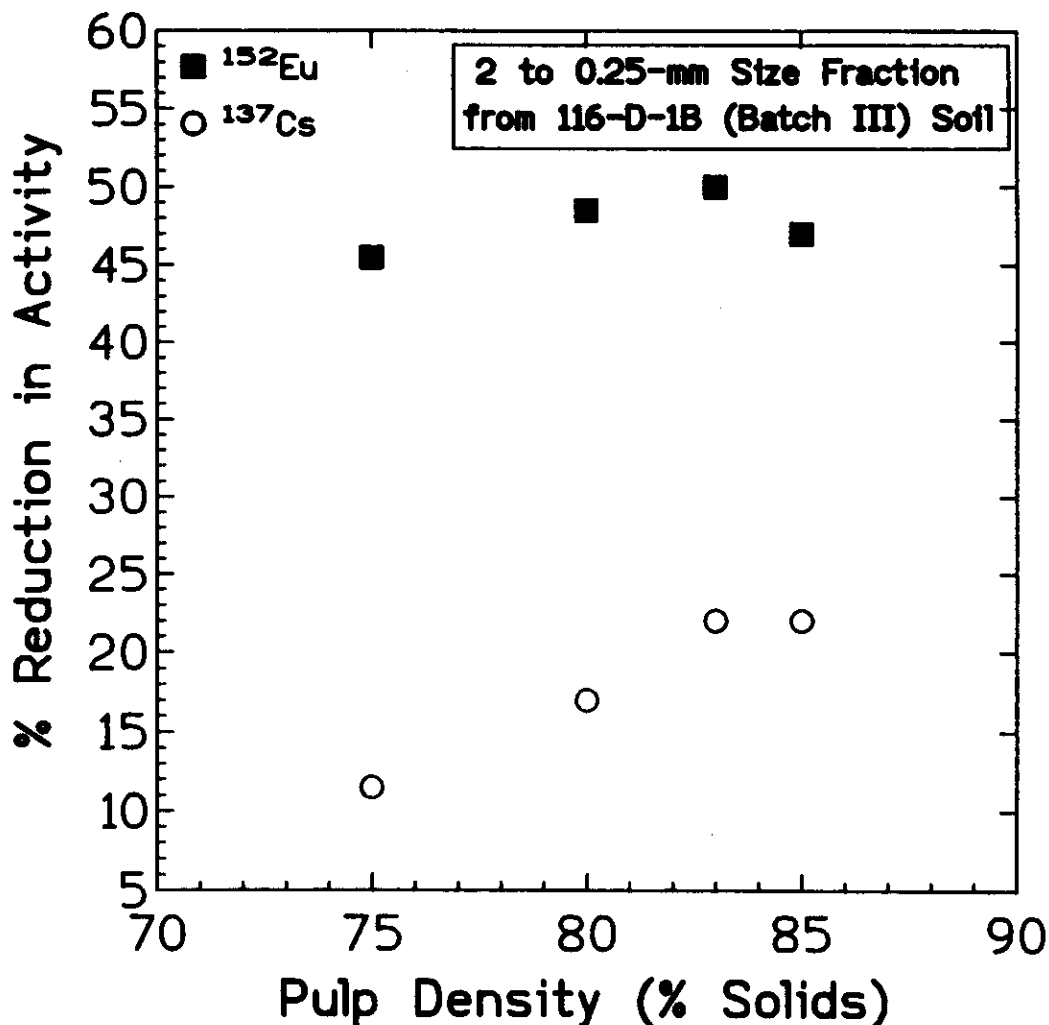
This very high selectivity for Cs by mica minerals in this soil appears to be the reason why  $^{137}\text{Cs}$  could not be removed to the same extent as  $^{60}\text{Co}$  and  $^{152}\text{Eu}$  through attrition scrubbing.

Table 6-1. Attrition Scrubbing Data for 2- to 0.25-mm-Sized Fractions of 116-D-1B Trench Soil: Relationship Between Pulp Density and Reduction in Radionuclide Activity.

Replicates	Pulp Density (Solids wt %)	Wt % Fines ( $<0.25$ mm) generated	Radionuclide Activities (pCi/g) in scrubbed fraction (2- to 0.25-mm)			Reduction (%) in Radionuclide Activity in scrubbed fraction (2- to 0.25-mm)		
			$^{60}\text{Co}$	$^{137}\text{Cs}$	$^{152}\text{Eu}$	$^{60}\text{Co}$	$^{137}\text{Cs}$	$^{152}\text{Eu}$
1	75	$<1$	1	81	18	67	10	53
2	75	1	$<1$	82	22	$>70$	9	42
3	75	4	1	77	22	67	14	42
4	75	2	$<1$	78	21	$>70$	13	45
1	80	2	$<1$	76	20	$>70$	16	47
2	80	1	$<1$	79	20	$>70$	12	47
3	80	4	$<1$	72	20	$>70$	20	47
4	80	3	$<1$	72	18	$>70$	20	53
1	83	4	$<1$	74	19	$>70$	18	50
2	83	4	$<1$	72	19	$>70$	20	50
3	83	4	1	70	20	67	22	47
4	83	4	$<1$	66	18	$>70$	27	53
1	85	3	$<1$	69	23	$>70$	23	39
2	85	2	$<1$	70	19	$>70$	22	50
3	85	-	$<1$	70	18	$>70$	22	53

Note: The activities of  $^{60}\text{Co}$ ,  $^{137}\text{Cs}$ , and  $^{152}\text{Eu}$  in wet-sieved, unscrubbed 2- to 0.25-mm- sized soil fractions were 3, 90, 38 pCi/g, respectively.

Figure 6-3. Percentage Reduction in Radionuclide Activity as a Function of Pulp Density.



Optical microscopic observations of scrubbed particles also indicated a more rounded appearance as compared to the angularity of unscrubbed material. Quantification by image analyses showed that scrubbing increased the quantities of finer material (i.e., increasing percentage of particles with smaller average equivalent diameters, (Figure 6-9). Scrubbing also produced increasing fractions of rounded particles (increasing roundness factor) by grinding up the more angular particles (Figure 6-10).

These experiments showed that  $^{60}\text{Co}$  and  $^{152}\text{Eu}$  activities in 2- to 0.25-mm-sized fractions of 116-D-1B soil were significantly reduced when attrition scrubbing was conducted at optimized pulp density of 83% (17% water) and at an energy input of 1.43 HP-min/lb. The residual activities of these two radionuclides in scrubbed soil were at or below the TPL. Another experiment was conducted to test the effects of using an electrolyte instead of water as the aqueous scrubbing medium (Table 6-3). These tests were conducted at the same

Table 6-2. Results From Attrition Scrubbing Tests on 116-D-1B Batch III Soil Sample:  
Reduction in Contaminant Radionuclide Activity as a Function of Input Energy

Replicates	Time (min)	Wt % fines (< 60 mesh) generated	HP	HP min/lb	Watt hr/lb	<sup>137</sup> Cs pCi/g	Cs % Reduction	<sup>152</sup> Eu pCi/g	Eu % Reduction	<sup>60</sup> Co pCi/g	Co % Reduction
1	5	4	0.059 2	0.27	3.36	72	20	22	42	1.4	53
2	5	4	0.060 3	0.27	3.36	74	18	19	50	1.3	57
1	10	7	0.070 6	0.64	7.96	66	27	19	50	0.7	77
2	10	6	0.052 7	0.48	5.97	71	21	19	50	<0.5	> 80
1	15	7	0.062 9	0.86	10.69	70	22	19	50	1.1	63
2	15	7	0.050 0	0.68	8.45	69	23	19	50	1.0	67
1	30	9	0.059 6	1.62	20.14	64	29	14	63	<0.5	> 80
2	30	9	0.045 4	1.24	15.42	67	26	16	58	<0.5	> 80
1	60	11	0.049 6	2.71	33.69	65	28	14	63	0.5	80
2	60	11	0.043 4	2.37	29.46	64	29	13	66	<0.5	> 80

\*Tests were conducted at 83% pulp density on 2- to 0.25-mm material. Initial activities (before scrubbing) of <sup>60</sup>Co and <sup>137</sup>Cs, and <sup>152</sup>Eu were 3, 90, and 38 pCi/g, respectively.



Figure 6-4. Percentage Reduction in Radionuclide Activity as a Function of Input Energy.

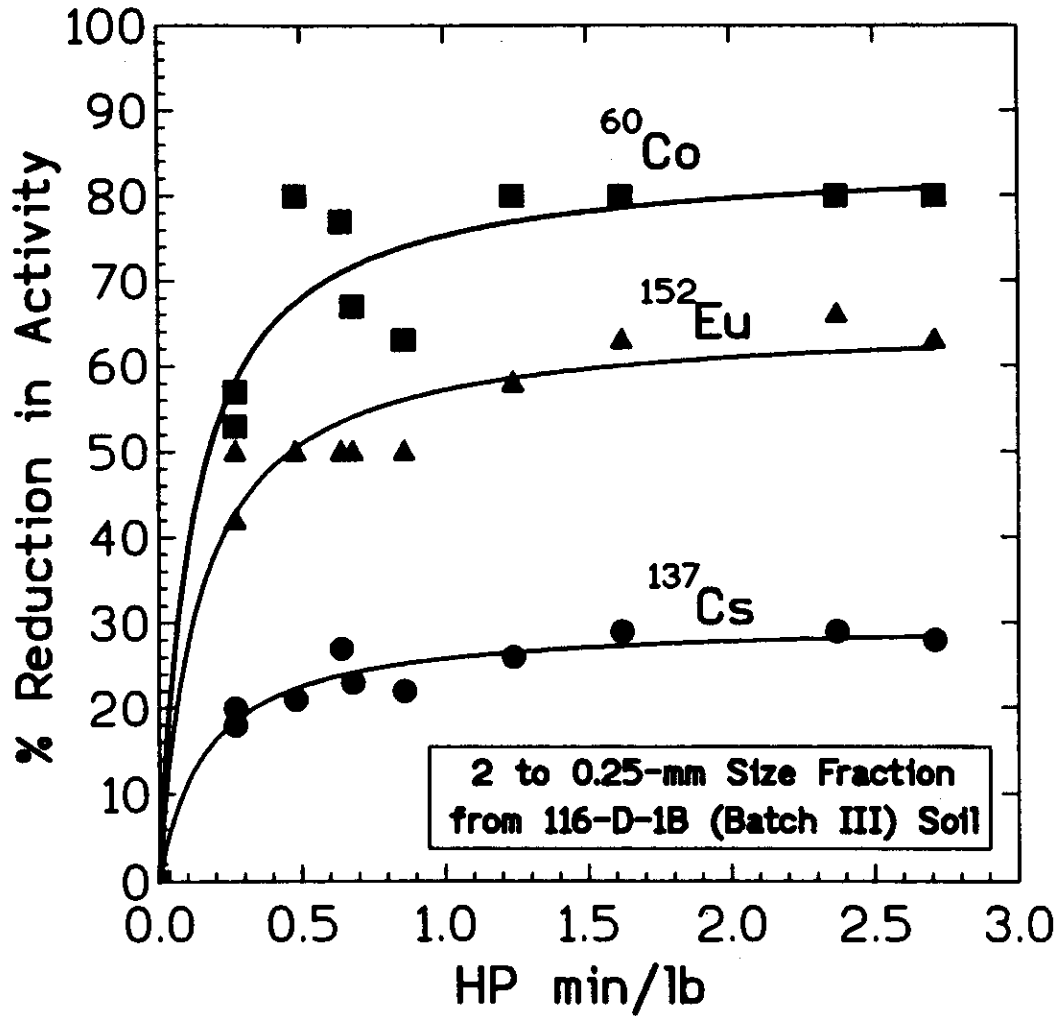


Figure 6-5. Percentage Fines generated During Attrition Scrubbing as a Function of Input Energy.

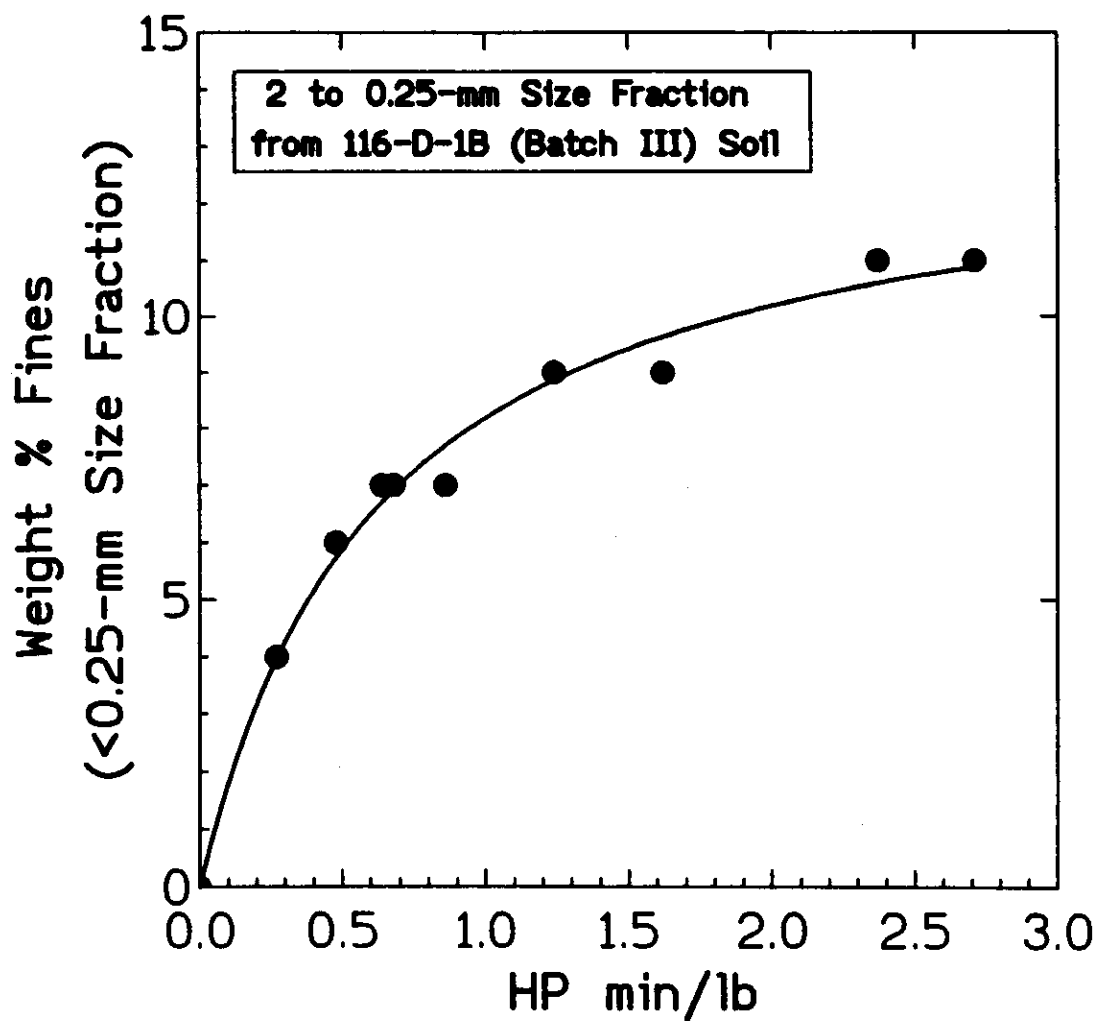


Figure 6-6. Observed Functional Relationships Between Impeller Torque and Time During Attrition Scrubbing Tests. The data represents duplicate tests.

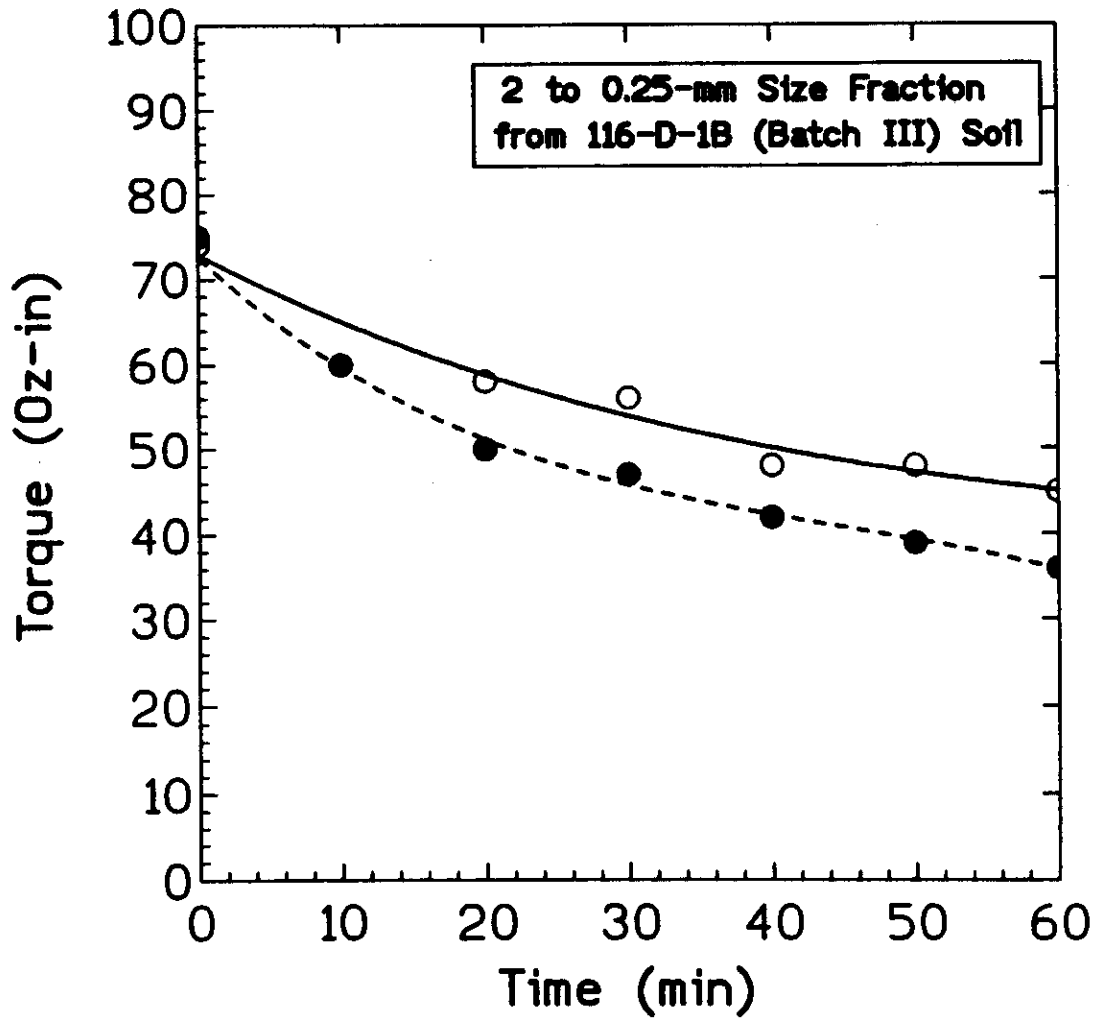


Figure 6-7. Optical Micrograph of Attritted Soil Particles from 116-D-1B Soil.

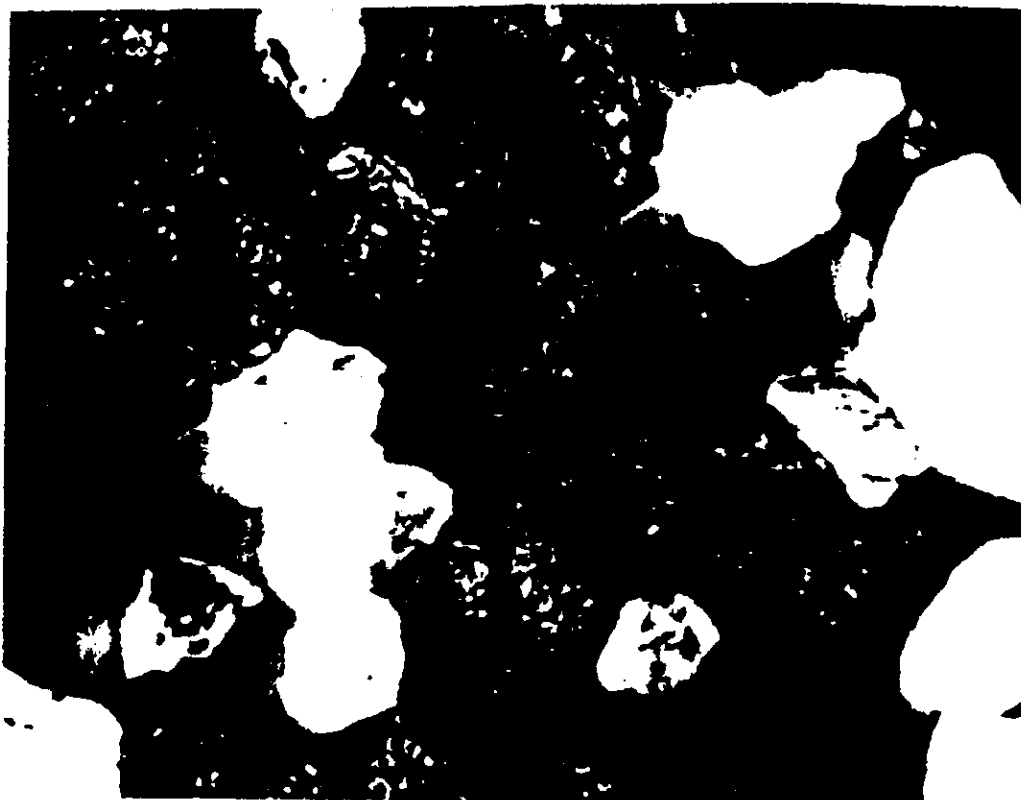


Figure 6-8. Scanning Electron Micrograph of a Mica Particle from 116-D-1B Soil.

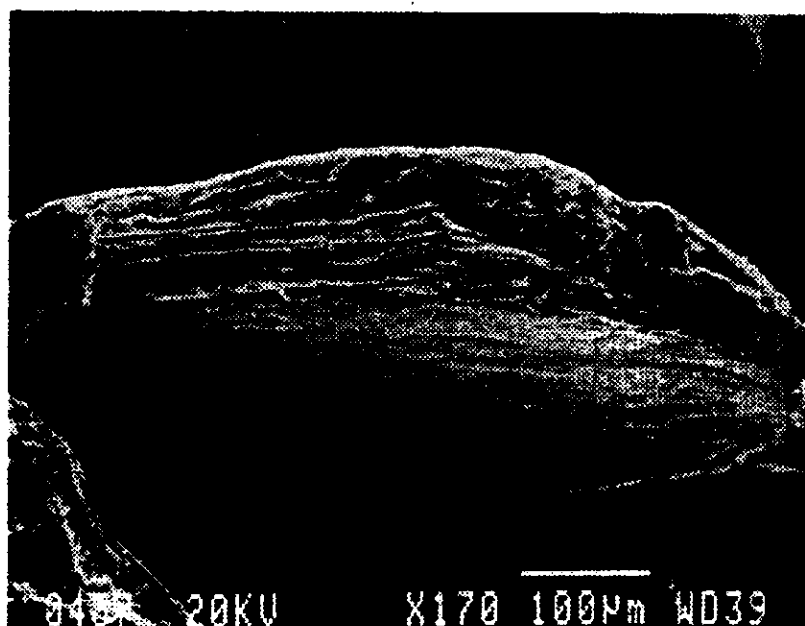


Figure 6-9. Effects of Attrition Scrubbing on Equivalent Particle Diameters.

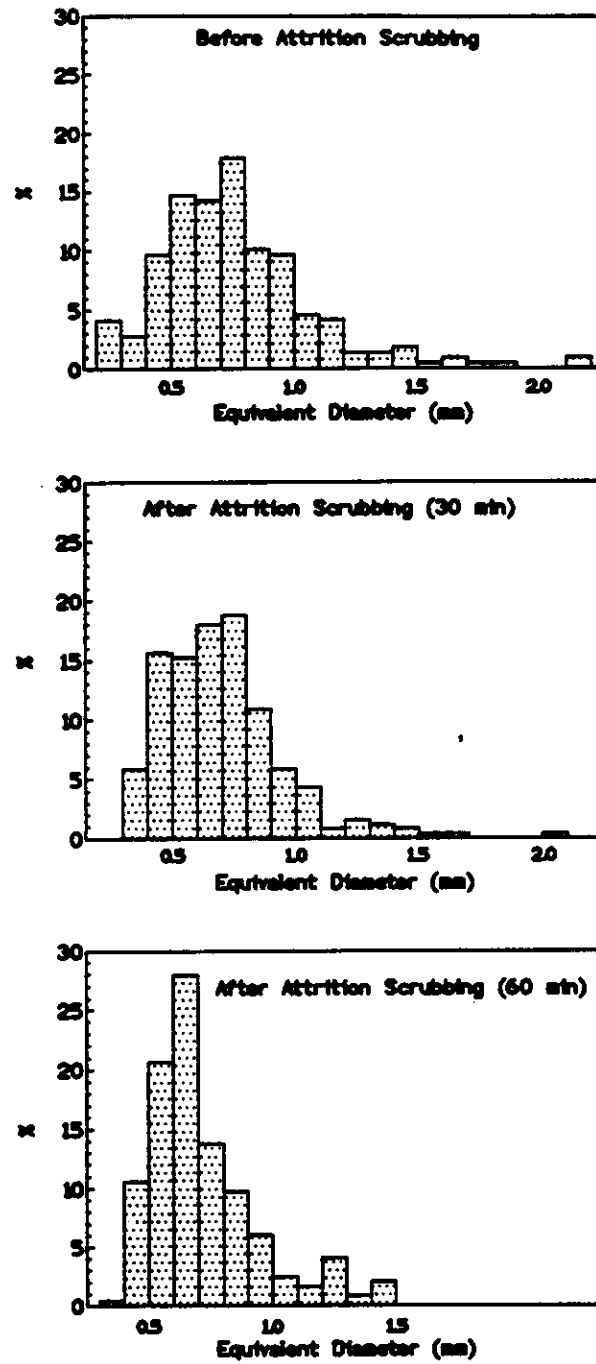


Figure 6-10. Effects of Attrition Scrubbing on Roundness Factor of Soil Particles.

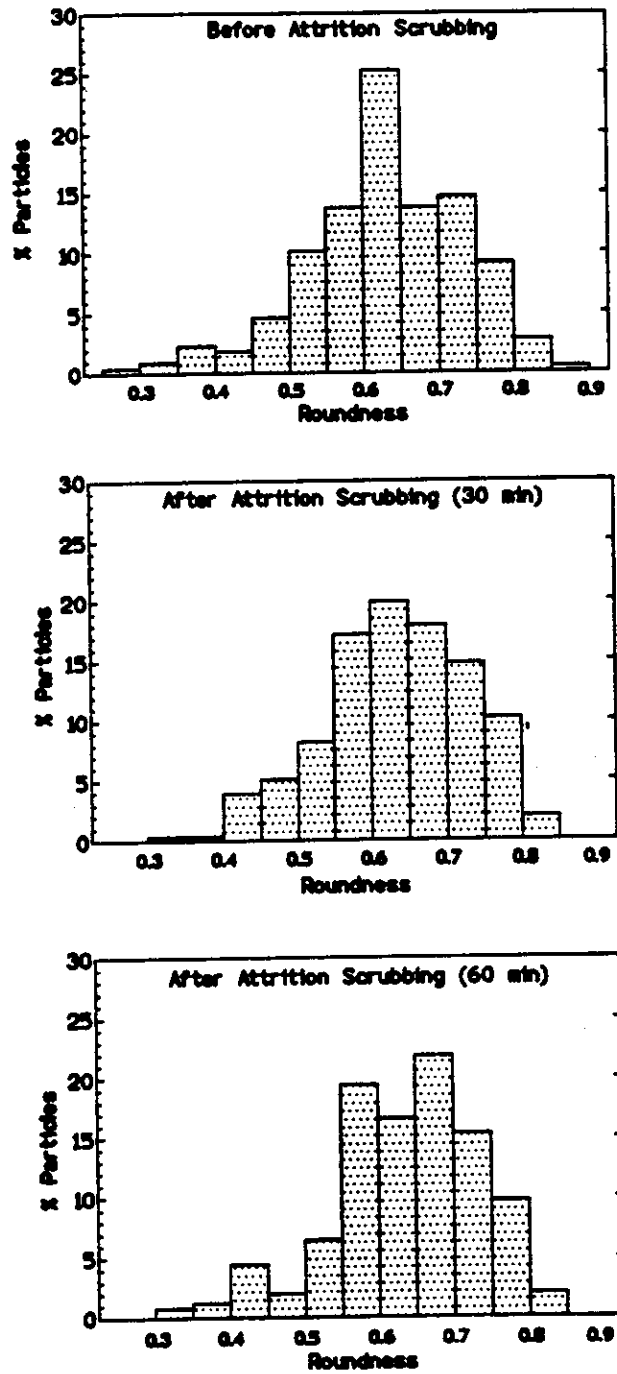


Table 6.3. Results of Optimized Attrition Scrubbing Tests with Water and Electrolyte on 2- to 0.25-mm-Sized Fractions of 116-D-1B Soil.

Replicate\Scrubbing Treatment	Pulp density %	Wt % fines < 60 mesh	Co % Reduction	Cs % Reduction	Eu % Reduction
1. Deionized water	83	9	> 80	29	63
2. Deionized water	83	9	> 80	26	58
1. Electrolyte	83	10	> 80	39	83
2. Electrolyte	83	10	> 80	39	83

\*The electrolyte solution consisted of 0.5 M ammonium citrate with pH adjusted to approximately 3 with citric acid.

optimum pulp density (83 %) and energy input (1.43 HP-min/lb) as the experiments conducted with water. The results indicated enhanced removal of  $^{137}\text{Cs}$  and  $^{152}\text{Eu}$  from the scrubbed material when electrolyte was used as the scrubbing medium. These tests with electrolyte generated approximately the same amount of fines (10% by weight) as the tests conducted with deionized water. Approximately > 80%  $^{60}\text{Co}$ , 39% of  $^{137}\text{Cs}$ , and 83% of  $^{152}\text{Eu}$  were removed from the soil by scrubbing with electrolyte. Such enhanced removal by electrolyte addition appears to be a result of the synergistic combination of scrubbing action, the improved dissolution of radionuclide-bearing surface coatings, and the reduced re-adsorption of solubilized contaminants onto freshly exposed surfaces of the coarse-grained soil.

Attrition experiments showed that under optimized conditions, scrubbing with water can reduce  $^{60}\text{Co}$  and  $^{152}\text{Eu}$  activities of the 2- to 0.25-mm-sized fraction of 116-D-1B soil to or below the TPL. Even though scrubbing with electrolyte improved the removal efficiency for all three radionuclides, none of these experiments could reduce the  $^{137}\text{Cs}$  activity in this soil fraction to values lower than its TPL.

## 7.0 AUTOGENOUS GRINDING

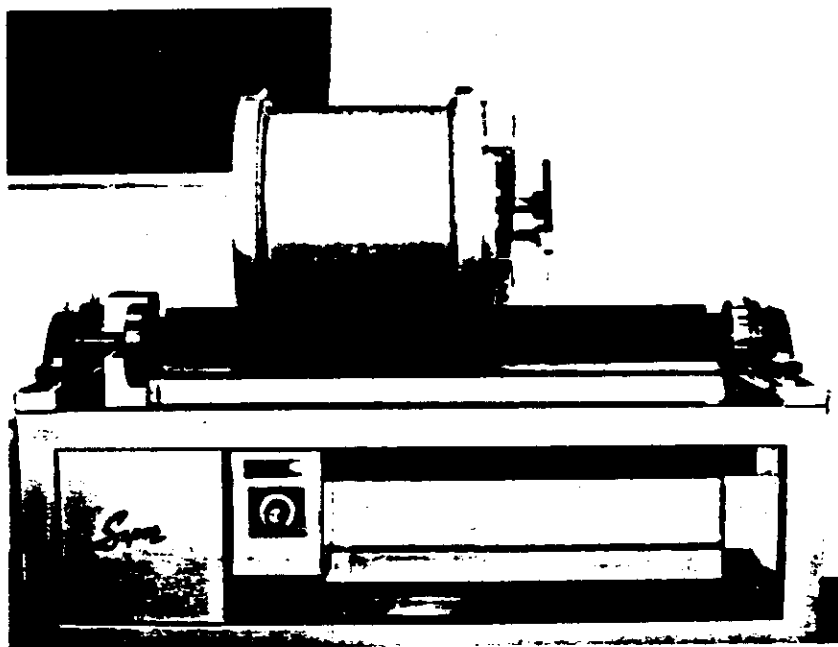
### 7.1 OBJECTIVE

The autogenous grinding experiments were conducted to test the removal of surface contamination from gravel- and cobble-sized material from 116-C-1 trench Batch II soils. The goal of this treatment was to remove surface contamination through the self-grinding action of cobbles and gravels. Typically, autogenous grinding is conducted using ball or rod mills (without steel balls or rods) running at speeds that promote crushing of the material. The objective of our experiments was to promote grinding of the contaminated particle surface and to minimize the pulverization of particles. The material from 116-C-1 trench Batch II consisted mainly (about 98% of total mass) of gravel-sized particles. Because washing this material with water did not significantly reduce the activity of radionuclides, additional physical treatment such as autogenous grinding was tested as a means to reduce the surface-bound contaminants.

### 7.2 EQUIPMENT AND PROCEDURES

The equipment used for autogenous grinding consisted of a Sepor 7.5-by-9-in. (internal dimensions) urethane-lined rod mill running on 2-in.-diameter, 24-in.-long neoprene rollers (Figure 7-1). This rod mill can grind 1 to 2.5 kg of material at speeds ranging from 20 to 320 RPM. Because the mill contains a rubber-gasketed lid, wet grinding could also be performed.

Figure 7-1. Laboratory-Scale Rod Mill Used for Autogenous Grinding.



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Autogenous grinding experiments were conducted on gravel-sized material obtained from 116-C-1 trench Batch II. All experiments were conducted on approximately 1-kg batches that were prewashed, air-dried, and counted for activities of  $^{60}\text{Co}$ ,  $^{137}\text{Cs}$ , and  $^{152}\text{Eu}$ . Grinding experiments were conducted under both dry and wet conditions. Additional experiments were conducted to measure any improvement in grinding performance that resulted from using some sand fraction (from 116-D-1B soil) as the abrasive material. To promote surface abrasion and minimize crushing of the material, all grinding experiments were conducted for 2 hours at 60% of the critical mill speed of  $114 \pm 5$  RPM.

### 7.3 RESULTS AND DISCUSSION

The data from autogenous grinding experiments are listed in Table 7-1. The dry or wet grinding (16% deionized water) experiments with and without sand were about equally effective in removing  $^{60}\text{Co}$ ,  $^{137}\text{Cs}$ , and  $^{152}\text{Eu}$  from the gravel-sized fraction of 116-C-1 Batch II material. Approximately, 74% to 85% of the  $^{60}\text{Co}$ , 14% to 17% of the  $^{137}\text{Cs}$ , and 89% to 97% of the  $^{152}\text{Eu}$  were removed using wet or dry grinding. However, dry grinding with or without sand was considered to be a nonviable treatment because such grinding generated extremely fine particles in the respirable range. Because of additional grinding action, wet grinding with sand removed about 7% more  $^{137}\text{Cs}$  and as much  $^{152}\text{Eu}$  and  $^{60}\text{Co}$  as wet grinding with water. However, adding sand to improve grinding performance resulted in a large mass (19%) of highly contaminated fines (<0.25-mm-sized fraction). A major fraction of fines (15%) generated in this experiment apparently resulted from ground-up sand. Generation of a large mass of contaminated fines during wet or dry grinding with sand, and production of soil particles in the respirable range, make these treatments less desirable than wet grinding without sand.

Another factor to note is that autogenous grinding is an energy-intensive process. For instance, the measured energy input (gross) to the rod mill ranged from 6.1 to 6.6 HP-min/lb. By comparison, the net energy used for actual grinding of the samples was computed to be about 0.3 HP-min/lb.

The results from these autogenous grinding experiments show that wet grinding with water was effective in reducing residual activities of  $^{60}\text{Co}$  and  $^{152}\text{Eu}$  in gravel fractions to much below the proposed TPL. To achieve similar cleanup levels for  $^{137}\text{Cs}$ , more than 97% of the initial activity needs to be removed from the gravel fractions. As none of these tests was effective in removing more than 21% of the initial  $^{137}\text{Cs}$  activity, it seems that autogenous grinding is not an effective process to reduce the residual activity of  $^{137}\text{Cs}$  in 116-C-1 Batch II soil material to below the proposed TPL.

Table 7-1. Autogenous Grinding Data for Gravel Fractions from 116-C-1 (Batch II) Soil\*.

Autogenous Grinding Treatment	<sup>137</sup> Cs (pCi/g)		% Red Cs Act.	<sup>152</sup> Eu (pCi/g)		% Red Eu Act.	<sup>60</sup> Co		% Red Co Act.	% Fines by wt
	Initial	Final		Initial	Final		Initial	Final		
16% DI water	822	707	14	69	2	97	16	2	85	0.5
Dry (25% sand**)	868	724	17	90	10	89	23	5	74	13.0***
17% DI water + 16.6% sand†	693	547	21	29	3	90	28	2	93	19.0††

\*Autogenous grinding was conducted at  $68 \pm 5$  RPM for 2 hours. Sand fraction (2 to 0.25 mm) from 116-D-1B soil was used as a grinding medium, with initial activities of <sup>60</sup>Co, <sup>137</sup>Cs and <sup>152</sup>Eu measured at 5, 95, and 47 pCi/g, respectively.

\*\*Final activities of <sup>60</sup>Co, <sup>137</sup>Cs, and <sup>152</sup>Eu in sand fraction were 6, 99, and 37 pCi/g, respectively.

\*\*\*Approximately 2% fines (<0.25 mm) were generated from the rocks, and 11% resulted from ground-up sand.

†Final activities of <sup>60</sup>Co, <sup>137</sup>Cs, and <sup>152</sup>Eu in sand fraction were 99, 822, and 374 pCi/g, respectively.

††Approximately 4% fines (<0.25 mm) were generated from the rocks, and 15% resulted from ground-up sand.

## 8.0 CHEMICAL EXTRACTION

### 8.1 OBJECTIVE

The objective of using chemical extraction was to evaluate the feasibility of removing contaminants from 116-C-1 (Batch II) and 116-D-1B (Batch III) trench soils using chemical solutions. This operation could be used if physical separation techniques do not remove contaminants to required levels or as a replacement to soil separation in the event that chemical extraction of the entire soil volume is more economical.

The chemical extraction of solids to selectively remove elements of interest is a proven technique used in the metallurgical and chemical processing industries for many years. The success of this technique generally lies in the proper selection of extractants (chemicals) and in understanding the kinetics of the reactions of concern. With this information, the proper selection of equipment to perform the extraction can be made and further scale-up studies can be conducted.

To process large quantities of soils, two main processing methods are available. The first choice is to use a stirred vat where contact of the leachant and soil can be easily controlled. The equipment for this process is relatively simple and can be scaled to handle very large volumes. The process can be operated in a continuous mode if a number of vats are used in series or operated in a batch mode with a single vat. The second possibility for leaching the soil is to add the extractant at the start of the soil-washing process (e.g., in the trommel) and to allow the reactions to take place while the physical separation is being performed. The advantage of this method is that very little equipment besides the soil-washing system is needed. However, the solids-to-liquid ratios and contact time in the trommel may not be optimal for the chemical extraction process to be effective.

A preliminary review of the literature was performed to identify candidate reagents for extracting soil to remove Co, Cs, and Eu. Extractants were selected to be tested both at ambient and elevated temperatures.

### 8.2 EQUIPMENT AND PROCEDURES

The extraction tests were performed using a 1-L capacity stirred vat system immersed in a temperature-controlled water bath. Tests were conducted by contacting 200 or 400 g of extractants with 100 g of 2- to 0.25-mm-sized fractions of 116-D-1B soil for a set period of time. Following extraction, the solids were separated and wet-sieved through a 0.25-mm sieve using deionized distilled water and oven dried at  $105 \pm 5^\circ\text{C}$ . The oven-dried solids and filtered liquids were analyzed for  $^{60}\text{Co}$ ,  $^{137}\text{Cs}$ , and  $^{152}\text{Eu}$ .

Extractants that were tested included acetic, citric, and hydrochloric acids that are typically used in chemical extractions of soils. Based on a review of soil chemistry literature, three widely used reductive extractants were also tested. One of these extractants consisted of a neutrally buffered solution (DCB) that contains sodium citrate and sodium bicarbonate with sodium dithionite added as a reductant (Jackson 1974). The extraction with this solution is conducted at about 80°C to prevent sulfur precipitation during extraction. The second reductive acid extractant consisted of a mixture of acetic acid and hydroxylamine hydrochloride (HAH) used as part of the sequential extraction procedure. The third extractant was a mixture of ammonium citrate and citric acid (CD) with sodium dithionite added as a reductant. Two additional extracts that were specifically formulated but are currently being evaluated for patents were also tested.

The effects of parameters such as extract composition, contact time, and temperature were investigated to optimize the extractive process for the 116-D-1B (Batch III) soil. Additionally, one of the currently proprietary extractants was used to statically extract (non-stirred system) the gravel fraction from 116-C-1 (Batch II) trench soil.

### 8.3 RESULTS AND DISCUSSION

The extraction data (Table 8-1) showed that all extractants except acetic acid removed substantial fractions of Co and Eu from the 2- to 0.25-mm-sized fractions of 116-D-1B (Batch III) trench soil. These extractants removed significantly more than the minimum of 40% Eu required for this soil to meet the TPL. These same extracts, however, showed significant differences in their ability to extract Cs. All the acid extractions conducted at room temperature removed only about 10% of Cs from soil. Standard reductive acid extractions (HAH and CD) extracted 20% to 34% more Cs than other acid extractions. Even though these extractions removed up to 44% of Cs, these removal efficiencies are below the minimum of 50% reduction necessary to meet the TPL for Cs. Therefore, none of these standard extractants can be used to reduce both Cs and Eu activities in the 2- to 0.25-mm-sized fraction adequately.

The data from specific extractions (Table 8-2) indicated that both the proprietary extractants (Extractant I and II) effectively removed major fractions of all three radionuclides from the 2- to 0.25-mm-sized fractions of 116-D-1B (Batch III) soil. When extractions were conducted for 6 hours at 96°C, both extractants removed 82% to 90% Co, 81% to 85% of Cs, and 64% to >99% of Eu from the soil. Reducing the extraction time in half did not significantly affect the fractions of radionuclides that were mobilized by Extractant II, indicating that the extractive chemical reactions were essentially complete at the end of 3 hours. When the extraction was conducted for 3 hours at a lower temperature (80°C), Extractant I removed about 10% less Cs and Eu but similar amounts of Co than the extraction conducted at 96°C.

Table 8-1. Standard Chemical Extraction Data for 116-D-1B Soil  
(2- to 0.25-mm-Sized Fraction)\*.

Radionuclide	Replicate	Extractants (% of Activity Removed)					
		0.5M HCl	0.5M Acetic Acid	0.5M Citric Acid	DCB	HAH	CD
<sup>60</sup> Co	1	80	20	83	>90	>90	>90
	2	>80	20	73	>90	>90	>90
<sup>137</sup> Cs	1	8	9	11	9	30	40
	2	11	11	11	11	30	44
<sup>152</sup> Eu	1	84	40	82	61	84	92
	2	87	40	82	61	84	92
Time (hr)		4	4	4	0.25	6	3
Temperature (°C)		25	25	25	80	96	80

\*The initial activities of <sup>60</sup>Co, <sup>137</sup>Cs, <sup>152</sup>Eu in the 2 to 0.25-mm-sized fraction of the soil were 3 to 5, 90 to 94, 38 to 52 pCi/g, respectively.

Table 8-2. Specific Chemical Extraction Data for 116-D-1B Soil  
(2- to 0.25-mm-Sized Fraction)\*.

Radionuclide	Extractants (% of Activity Removed)					
	Ext I-A	Ext I-B	Ext II-A	Ext II-B	Ext II-C	Ext II-D
<sup>60</sup> Co	>90	82	>90	>90	>90	>90
<sup>137</sup> Cs	69	81	85	85	85	84
<sup>152</sup> Eu	55	64	>99	>99	>99	>99
Time (hr)	3	6	3	4	6	6
Temperature (°C)	80	95	95	95	95	95

\*The initial activities of <sup>60</sup>Co, <sup>137</sup>Cs, and <sup>152</sup>Eu in the 2- to 0.25-mm-sized fraction of the soil were 3 to 5, 90 to 94, and 38 to 52, respectively. The solid solution ratio in all experiments except Ext II-3 were 1:4. Ext II-3 used a 1:2 ratio.

The effects of reduced chemical concentrations of Extractant II on the extractive efficiency are listed in Table 8-3. The data show that when the concentration of the major chemical component is decreased to 25% of the initial concentration (Extractant II-1, 2), about 70% (15% less than the full strength extraction) of Cs was extracted. The reduced extract concentration, however, did not affect the extractability of Co and Eu. Changing the

solid: solution ratio of extraction from 1:4 to 1:2 did not change the amounts of Co and Eu that were extracted (Extractant II-3), but extracted only about 57% of Cs in the soil. The last two experiments conducted with the major extractant component's concentration at 5% of the original strength showed that even at these low concentrations, the extractant released about 90% each of Co and Eu and about 56% of Cs. Therefore, the use of the lowest strength Extractant II will remove sufficient quantities of all three radionuclides from the 116-D-1B composite soil fraction to meet the proposed TPL. Moreover, if chemical extraction is selected as part of the soil-washing process for this soil, use of the lowest strength Extractant II will significantly reduce the cost of the extractive step.

Based on the chemical extraction data for 116-D-1B (Batch III) soil, all static chemical leaching of gravel fractions of 116-C-1 (Batch II) soil was conducted with the most effective reagent, Extractant II. The results of these experiments (Table 8-4) indicated that leaching for 6 hours either at ambient temperature or 96°C reduced the residual activities of Co and Eu in the gravel fraction to less than the proposed TPL. Despite achieving up to 40% removal by static leaching at 96°C, the residual Cs activity in the gravel fraction was at least an order of magnitude (> 350 pCi/g) greater than the TPL of 30 pCi/g.

The chemical extraction experiments showed that it is possible to reduce the activities of all three radionuclides in the 116-D-1B soil to less than the proposed TPL. Static chemical leaching was not as effective for treating gravel fractions from the 116-C-1 Batch II soils because of inherently higher Cs activities in this sample.

Table 8-3. Effects of Specific Extractant Concentration and Solid/Extractant Ratio on Contaminant Radionuclide Removal from 116-D-1B Soil (2- to 0.25-mm-Sized Fraction)\*.

Radionuclide	Extractions (% Activity Removed)				
	Ext II-1	Ext II-2	Ext II-3	Ext II-4	Ext II-5
<sup>60</sup> Co	88	92	91	95	84
<sup>137</sup> Cs	73	67	57	55	57
<sup>152</sup> Eu	>99	97	97	91	90
Time (hr)	6	6	6	6	6
Temperature (°C)	95	95	95	95	95

\*The initial activities of <sup>60</sup>Co, <sup>137</sup>Cs, and <sup>152</sup>Eu in the 2- to 0.25-mm-sized fraction of the soil were 3 to 5, 90 to 94, and 38 to 52 pCi/g, respectively. The solid solution ratio in all experiments except Ext II-3 were 1:4. Ext II-3 used a 1:2 ratio.

Table 8-4. Specific Chemical Extraction Data for 116-D-1B Soil.

Extraction Temp. °C	Before Leaching (pCi/g)			After Leaching (pCi/g)			Reduction in Activity (%)		
	<sup>137</sup> Cs	<sup>152</sup> Eu	<sup>60</sup> Co	<sup>137</sup> Cs	<sup>152</sup> Eu	<sup>60</sup> Co	<sup>137</sup> Cs	<sup>152</sup> Eu	<sup>60</sup> Co
1. 96	1046	30	11	584	3	2	44	90	82
2. 96	587	28	11	376	2	2	36	93	82
1. 24	424	24	9	365	11	5	14	54	44

\*All leaching was conducted with Extractant II under static conditions for 6 hours.

## 9.0 COMBINATION TESTS

### 9.1 OBJECTIVE

The goal of these experiments was to determine whether contaminant radionuclides can be mobilized to a greater degree by combining different treatments. For 116-D-1B soil, these tests consisted of combinations of two successive attrition scrubblings using water or an electrolyte. The electrolyte was used to examine if radionuclide release can be enhanced through a combination of chemical leaching action and attrition scrubbing and also if re-adsorption of released contaminants onto the cleaned soil substrate can be attenuated through the use of a competitive adsorbing cation in the electrolyte. Similarly, a test that combined autogenous grinding in the presence of Extractant II was performed on the gravel-sized fraction of the 116-C-1 Batch II soil to examine if increasing quantities of contaminants can be released through the combined action of surface grinding and chemical leaching.

### 9.2 DESCRIPTION OF TESTS

The attrition scrubbing tests were conducted with the equipment that was described previously (Section 6.2). All scrubbing tests were conducted on approximately 500-g loads of previously wet-sieved and air-dried 2- to 0.25-mm-sized fractions of 116-D-1B trench soil. Based on the optimum performance data obtained from previous scrubbing experiments, all tests were conducted with pulp densities set at 83%. The electrolyte solution consisted of 0.3M ammonium citrate acidified to near pH 3 with citric acid. The scrubbing was conducted at 900 RPM for residence times of 30 min (energy input of 1.43 HP-min/lb). The two-stage scrubbing tests were performed by conducting the first-stage scrubbing with water or electrolyte, and wet-sieving the scrubbed fractions through a 0.25-mm sieve to remove the fines that were generated during scrubbing. Removal of fines after scrubbing was necessary because previous data (Section 6.3) showed that accumulation of fines reduced the scrubbing efficiency. The second-stage attrition scrubbing was conducted on the washed and air-dried 2- to 0.25-mm-sized material, with water or electrolyte added to bring the pulp densities up to 83%. After completing the second-stage scrubbing, the material was again wet-sieved through a 0.25-mm sieve and dried at  $105 \pm 5^\circ\text{C}$ , and representative subsamples were taken for radionuclide analyses ( $^{60}\text{Co}$ ,  $^{137}\text{Cs}$ , and  $^{152}\text{Eu}$ ).

Two combination tests on gravel-sized fractions from 116-C-1 trench soil involving autogenous grinding were conducted using the equipment described previously in Section 7.2. All grinding experiments were conducted at milling speeds of  $68 \pm 5$  RPM. The combination experiment was conducted on approximately 1-kg batches that were prewashed, air dried, and counted for activities of  $^{60}\text{Co}$ ,  $^{137}\text{Cs}$ , and  $^{152}\text{Eu}$ . The first experiment involved an autogenous grinding for 2 hours in an electrolyte solution. The goal of this experiment was to reduce any potential re-adsorption of released contaminants onto the freshly ground surface of the gravel. The composition of the electrolyte used in this experiment was the same as



that used in the combination experiment involving the 116-D-1B soil (0.3M ammonium citrate, pH 3 with citric acid). The second experiment consisted of surface-grinding the gravel-sized fraction for 6 hours with sufficient quantity of Extractant II added to bring the solid concentration up to 50% by weight. At the conclusion of these experiments, the fines were washed off the gravel fractions with water, the coarse fractions were oven dried, and samples were drawn for counting the residual activities of contaminant radionuclides.

### 9.3 RESULTS AND DISCUSSION

The combination test data (Table 9-1) indicated that attrition scrubbing conducted in two stages with water on average removed 34% of Cs and 71% of Eu from the 2- to 0.25-mm-sized fractions of 116-D-1B soil. No differences were observed in the removal of Co, as >80% of this contaminant was released in all cases. As compared to the single-stage scrubbing data, two-stage scrubbing removed an additional 6% of Cs and 10% of Eu, and generated about 4% more (total 13%) fines from the soil (see Tables 6-3 and 9-1). Similar improvement in Cs and Eu removal was observed when two-stage scrubbing was conducted with electrolyte. Two-stage scrubbing with electrolyte removed about 10% more Cs and Eu, and produced only an additional 3% fines as compared to the once-through scrubbing with electrolyte (see Tables 6-3 and 9-1). An evaluation of the two-stage scrubbing performance with water and electrolyte indicated that the highest quantities of radionuclide removal (about 48%  $^{137}\text{Cs}$ , and 94%  $^{152}\text{Eu}$ ) were achieved when scrubbing was conducted with the electrolyte. These removal rates were about 14% more for Cs and 23% greater for Eu than the reduction efficiencies achieved by scrubbing with water only. Radionuclide removals were improved by generating the same amount of fines (about 13%), indicating that even though the scrubbing intensities (approximately the same energy input in each stage) were the same in both experiments, the enhanced radionuclide removal with electrolyte was probably due to the more intense extractive action and blocking of re-adsorption of mobilized radionuclides onto the soil mineral surfaces.

The choice of electrolyte to achieve this goal was based mainly on the mineralogical and sequential extract data. The electrolyte mixture used in this experiment contained ammonium ions, that are known to compete effectively with Cs for adsorption and wedge sites on minerals and sufficient proton activity (acidity) to enhance dissolution of contaminant-bearing hydrous oxide phases on mineral surfaces. This combination test proved that attrition scrubbing with an appropriately formulated electrolyte solution can synergistically increase the contaminant mobilization from this soil. The fractions of contaminants removed (>80%  $^{60}\text{Co}$ , and about 48%  $^{137}\text{Cs}$ , and 94%  $^{152}\text{Eu}$  using the combination test indicated that two-stage attrition scrubbing with the electrolyte provides another technique (in addition to chemical extraction) to reduce the activities of these radionuclides in 116-D-1B soil composite to less than the proposed TPL. The choice of appropriate treatment technique for this soil would be based on ease of application and cost.

The results of the combination test involving autogenous grinding in the electrolyte and in the Extractant II (Table 9-2) showed that both combination tests were equally effective

for removing major fractions of Co (88% to 95%) and Eu (94% to 97%) from the gravel-sized fraction of 116-C-1 (Batch II) soil. However, similar amounts of activities (85% Co and 97% Eu) were removed when grinding was conducted using deionized distilled water (see Table 7-1). These data indicate that, once mobilized, these two radionuclides apparently do not re-adsorb onto the gravel surfaces. Contrary to the behavior of Co and Eu, mobilization of Cs was dependent on the type of solution used with grinding. Grinding with Extractant II released the greatest amount of Cs (53%), whereas grinding with electrolyte released only 25% Cs (see Table 9-2). Both these combination tests released greater fractions of Cs as compared to the smaller Cs fraction (14%) mobilized when grinding was conducted with water. These data showed that Cs is much harder to mobilize than either Co or Eu and that re-adsorption of released Cs is a significant phenomena in these soils. These combination tests showed that despite significant increases in Cs release, the residual activity of this radionuclide in the gravel-sized fractions remained at least an order of magnitude greater than the proposed TPL.

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Table 9-1. Results from Two-Stage Attrition Scrubbing Tests in Water or in an Electrolyte Conducted on Sand Fractions (2 to 0.25 mm Size) from 116-D-1B Soil\*.

Replicate/Scrubbing Treatment	Wt % fines < 60 mesh	<sup>60</sup> Co (pCi/g)	<sup>137</sup> Cs (pCi/g)	<sup>152</sup> Eu (pCi/g)	Co % Reduction	Cs % Reduction	Eu % Reduction
1. Two-stage in DI water	12	< 1	63	15	> 80	33	71
2. Two-stage in DI water	13	< 1	62	15	> 80	34	71
1. Two-stage in electrolyte	12	< 1	48	1.6	> 80	49	97
2. Two-stage in electrolyte	13	< 1	50	5	> 80	47	90

\*Measured activities of <sup>60</sup>Co, <sup>137</sup>Cs, and <sup>152</sup>Eu in 2- to 0.25-mm-sized fraction were 4.8, 94, and 52 pCi/g, respectively. The electrolyte solution consisted of 0.5M ammonium citrate with pH adjusted to 3 with citric acid.

Table 9-2. Results from Autogenous Grinding Tests with an Electrolyte or a Specific Extractant Conducted on Gravel-Sized Fractions from 116-C-1 Batch II Soil.

Autogenous Grinding Treatment	<sup>60</sup> Co (pCi/g)		% Red Co Act.	<sup>137</sup> Cs (pCi/g)		% Red Cs Act.	<sup>152</sup> Eu (pCi/g)		% Red Eu Act.	% Fines by wt
	Initial	Final		Initial	Final		Initial	Final		
With 17% electrolyte	26	3	88	883	665	25	88	5	94	5.0
With 50% Extractant II	19	1	95	751	351	53	55	2	97	7.3

## 10.0 WASTE WATER TREATMENT

### 10.1 OBJECTIVE

The overall cost and feasibility of using soil-washing techniques for the treatment of radionuclide-contaminated soils depend partly on the ability to treat the liquid wastes generated. The liquid waste streams may include water from water-washing operations used in the physical separation process (e.g. grizzly, trommel, screens, hydrocyclone), spent chemical solutions from chemical extraction operations (leachates for recycle or disposal), or water or electrolytes from attrition scrubbing operations.

One of the goals for developing an optimum soil-washing flow sheet is to minimize waste stream volume. Achievement of this goal requires that much of the water and chemical solutions be recycled. The concentrations of suspended solids in recycled water and solutions should be reduced to levels low enough to prevent detrimental impact on processing equipment such as spray nozzles and pumps. Typically, the suspended solid load in waste streams is reduced through settling and, if necessary, through flocculation. Additional processing of the streams may also be necessary to remove dissolved contaminant buildup in the recycle and bleed streams.

The objective of our water tests was to evaluate the feasibility of removing suspended soil particles from liquid streams generated during bench-scale soil-washing tests conducted on 100-D-1B (Batch III) soil. These waste streams included wash water generated during wet-sieving, two-stage attrition scrubbing with an electrolyte, and chemical extraction. A set of flocculation tests was conducted on each of these waste streams. Measured parameters included turbidity, pH, temperature, and activities of selected radionuclides remaining in clarified liquids.

### 10.2 EQUIPMENT AND PROCEDURES

The flocculation tests were conducted with a 6-station Phipps and Bird batch reactor unit equipped with stainless-steel stir paddles. Turbidity measurements were conducted using a Hach Ratio/XR Turbidity meter calibrated with formazine standards. The waste solutions used in these tests were generated during soil-washing feasibility experiments conducted on 116-D-1B (Batch III) soil. These aqueous streams consisted of wash water generated from wet-screening of <2-mm soil material over a <0.25-mm (60-mesh) sieve, electrolyte containing wash solution resulting from two-stage attrition scrubbing of 2- to 0.25-mm fraction, and spent solution resulting from chemical extraction (with optimum extractant II)

of 2- to 0.25-mm size fraction material. All waste solutions were mixed thoroughly and allowed to settle for 15 min to remove larger particles. Six 250-mL aliquots were then removed from each of the waste solutions and placed in 600-mL beakers.

Two polymeric flocculation agents were used in these experiments. One of these consisted of a cationic polymer CATFLOC-L<sup>®</sup>, a primary coagulant. This coagulant was chosen because it is effective over a broad pH range, whereas conventional flocculents such as ferric chloride or alum are effective over a relatively narrow pH range. Because of its effectiveness, CATFLOC-L<sup>®</sup> has been used to treat actinide-containing pond waters from Rocky Flats (Triay et al. 1993). The second flocculent was a high-molecular weight anionic polymer (POL-E-Z-692<sup>®</sup>) that is effective at low pH values, promotes floc stability, and improves filterability of treated solutions. Only CATFLOC-L<sup>®</sup> was used to treat the wash water from wet sieving and the spent chemical extractant solutions. Both CATFLOC-L<sup>®</sup> and POL-E-Z-692<sup>®</sup> were used to treat the highly turbid waste solutions generated from two-stage attrition scrubbing tests.

CATFLOC-L<sup>®</sup> was added to the stirred waste solutions (wet-screening waste waters, and spent chemical extractant) at concentrations of 0.5, 5, 10, 15, and 30 mg/L. The waste water in the sixth beaker in each set was used as the control solution where no flocculent was used. The attrition scrubbing waste solution was treated with POL-E-Z-692<sup>®</sup> (0.5, 5, 19, 15, and 30 mg/L) and CATFLOC-L<sup>®</sup> at a fixed concentration of 10 mg/L. All solutions were stirred at 85 RPM for 10 to rapidly mix the flocculents with the suspended solids. Then the solutions were agitated for 20 min at a stirrer speed of 25 RPM. Next, the stirrers were turned off and the flocs were allowed to settle for a period of 30 min.

After the 30-min settling period, 100-mL aliquots were drawn off from the upper part of solution in each 600-mL beaker and the temperature was measured. The remaining solutions in the 600-mL beakers and the 100-mL aliquots were both allowed to settle for an additional 24 hr. First, turbidity measurements were conducted on 30 mL of clear supernatant from each 100-mL aliquot. These measurements represented turbidities after 24 hr settling time. After these measurements were completed, the clear solutions were mixed back with the remaining 70 mL of aliquots, and each 100 mL aliquot was agitated and the turbidity of dispersed solution was measured. The latter measurements represented 30 min settling-time turbidities. After the 24-hr settling time additional clear supernatant was drawn off each 600-mL beaker to measure the radionuclide activities in solution.

### 10.3 RESULTS AND DISCUSSION

The flocculation tests showed that CATFLOC-L<sup>®</sup> was an effective flocculent for wash water generated from wet-sieving (Table 10-1). After 24 hr of settling, turbidity values of <4 nephelometric turbidity units (NTUs) were achieved for flocculent addition levels of

≥5 mg/L. The flocculent additions did not significantly alter the pH of waste water. The radionuclide activities in flocculated supernatants were below the minimum detectable activities (MDAs) for all three radionuclides ( $^{60}\text{Co}$ : <1.7 pCi/mL;  $^{137}\text{Cs}$ : <1.6 pCi/mL; and  $^{152}\text{Eu}$ : <4.3 pCi/mL). Therefore, using CATFLOC-L<sup>®</sup> at a concentration of 5 mg/L, it is possible to remove the bulk of the suspended solids from these wash waters.

Significant reductions in turbidity were also observed when the waste water from two-stage attrition scrubbing with electrolyte was treated with a combination of the flocculents CATFLOC-L<sup>®</sup> and POL-E-Z 692<sup>®</sup>. Turbidity values of ≤90 NTU were observed at POL-E-Z 692<sup>®</sup> concentrations of ≥5 mg/L. The activities of all radionuclides were close to or below MDA in both treated and untreated two-stage attrition scrubbing waste solutions. Use of flocculents did not affect the pH of this waste water. Compared to the untreated waste solution, flocculent treated solutions showed maximum turbidity reductions of 95 % after 30 min settling, and 80 % at the end of 24 hr. These tests showed that it is feasible to flocculate a major fraction of colloidal particles from the highly turbid waste solutions resulting from two-stage scrubbing with an electrolyte.

The spent solution from chemical extraction was representative of very low turbidity (350 NTU at 30 min, and 35 NTU at 24 hr) wastes. Therefore, addition of the flocculent (CATFLOC-L<sup>®</sup>) resulted in a maximum reduction in turbidities of only 57 % and 31 % at the end of the 30-min and 24-hr settling times, respectively. The pH of the spent solution was not significantly affected by the flocculent additions. The activities of both  $^{60}\text{Co}$  and  $^{152}\text{Eu}$  in this waste water before or after treatment were below MDA. The activity of  $^{137}\text{Cs}$  in the supernatant after 24 hr settling with or without flocculent was approximately 6 pCi/mL, indicating that this radionuclide was present mainly as a dissolved constituent. This test showed that a small addition of CATFLOC-L<sup>®</sup> (0.5 to 30 mg/L) will slightly improve the settling rate of the suspended solids after 30 min, but little settling improvement is noted after 24 hr. The removal of dissolved  $^{137}\text{Cs}$  will require additional water treatment unit operations such as ion-exchange or precipitation.

These flocculation tests showed that it is feasible to remove suspended solids thorough additions of polyelectrolytes (CATFLOC-L<sup>®</sup>, and POL-E-Z 692<sup>®</sup>). The activities of radionuclides ( $^{60}\text{Co}$  and  $^{152}\text{Eu}$ ) were close to or below MDA in supernatants of all treated and untreated waste waters. As expected, the activity of  $^{137}\text{Cs}$  in spent solutions from chemical extraction was higher (about 6 pCi/L) than in the other two waste streams because chemical extraction removed more cesium from 2 to 0.25-mm fraction of 116-D-1B (Batch III) soil.

These tests indicated that the suspended solids in liquid waste streams generated from soil-washing of 116-D-1B (Batch III) soil can be effectively flocculated using commercially available polyelectrolytes. However, additional factors that need to be examined include the

effects of recycling effluent on the build-up of soluble contaminants and macroions and the use of ion-exchange or precipitation processes for radionuclide removal from solution. Future tests should also include optimizing (i.e. minimizing) the amounts of polyelectrolyte flocculents needed to achieve desired settling rates.

Table 10-1. Results of Flocculation Experiments on Waste Waters from 116-D-1B (Batch III) Soil-Washing Tests

Flocculent Concentration (mg/L)	Temperature °C	pH	Turbidity (30 min) NTU <sup>(a)</sup>	Turbidity (24 hr) NTU	Radionuclide Activity in Supernatant (24 hr) pCi/mL		
					<sup>60</sup> Co	<sup>137</sup> Cs	<sup>152</sup> Eu
Wash Water from Wet-Sieving							
No Flocculent	25.5	8.0	1100	50	<1.7	<1.6	<4.3
CATFLOC-L <sup>o</sup> (0.5)	25.0	8.2	800	45	<1.7	<1.6	<4.3
CATFLOC-L <sup>o</sup> (5)	24.5	8.2	330	3.2	<1.7	<1.6	<4.3
CATFLOC-L <sup>o</sup> (10)	24.5	7.8	39	3.1	<1.7	<1.6	<4.3
CATFLOC-L <sup>o</sup> (15)	24.0	7.7	60	1.5	<1.7	<1.6	<4.3
CATFLOC-L <sup>o</sup> (30)	25.0	7.7	210	2.7	<1.7	<1.6	<4.3
Waste Water from Two-Stage Attrition Scrubbing with Electrolyte							
No Flocculent	23.1	3.6	8400	350	<1.7	<1.6	5.1
CATFLOC-L <sup>o</sup> (10) + POL-E-Z 692 <sup>o</sup> (0.5)	22.7	3.6	2400	240	<1.7	1.8	5.2
CATFLOC3.6-L (10) + POL-E-Z 692 <sup>o</sup> (5)	22.6	3.6	500	90	<1.7	<1.6	6.1
CATFLOC-L <sup>o</sup> (10) + POL-E-Z 692 <sup>o</sup> (10)	22.6	3.6	400	70	<1.7	2.5	<4.3
CATFLOC-L <sup>o</sup> (10) + POL-E-Z 692 <sup>o</sup> (15)	22.4	3.6	450	70	<1.7	<1.6	<4.3
CATFLOC-L <sup>o</sup> (10) + POL-E-Z 692 <sup>o</sup> (30)	22.6	3.6	800	80	<1.7	<1.6	<4.3
Spent Solution from Chemical Extraction with Extract II							
No Flocculent	23.5	2.2	350	35	<1.7	5.5	<4.3
CATFLOC-L <sup>o</sup> (0.5)	23.1	2.2	160	24	<1.7	6.5	<4.3
CATFLOC-L <sup>o</sup> (5)	23.0	2.2	150	31	<1.7	5.7	<4.3
CATFLOC-L <sup>o</sup> (10)	23.1	2.3	170	32	<1.7	5.9	<4.3
CATFLOC-L <sup>o</sup> (15)	22.8	2.3	170	32	<1.7	6.1	<4.3
CATFLOC-L <sup>o</sup> (30)	23.2	2.3	180	41	<1.7	5.1	<4.3

(a) NTU = nephelometric turbidity unit.



## 11.0 DISCUSSION AND RECOMMENDATIONS

### 11.1 CONTAMINANT RADIONUCLIDES IN 100 AREA SOILS

The radionuclide data indicated that the principal contaminants in the bulk soils (116-C-1, Batch I, II; and 116-D-1B, Batch III) were  $^{137}\text{Cs}$ ,  $^{60}\text{Co}$ , and  $^{152}\text{Eu}$ . Chemically these three radionuclides belong to three different groups. Therefore, they exhibit distinctly different geochemical behavior.

Cesium is an alkali element and its chemical properties are similar to other elements of this group (Li, Na, K, Rb, and Na). All alkali elements are univalent cations and their adsorption preference on mineral surfaces increases with increasing ionic radii ( $\text{Li} < \text{Na} < \text{K} < \text{Rb} < \text{Cs}$ ). Because of larger ionic radius and less degree of hydration, Cs adsorbs with higher affinity than other alkali cations. Cesium ions are known to adsorb specifically on wedge sites of micas where they can substitute for the potassium ions that commonly occupy these interlayer sites. These specifically adsorbed cesium ions are hard to displace by any other cations except those with comparable ionic radii and hydration status. Ammonium ions due their ionic radii being similar to cesium can effectively compete with cesium for the highly specific wedge sites. The data obtained in our experiments confirmed that attrition scrubbing of 116-D-1B (Batch III) soil with ammonium ion- containing electrolyte released more cesium than that released when scrubbing was conducted with deionized water. This observation is also supported by the enhanced removal of cesium achieved by the acid-ammonium acetate extraction as compared to the other extractive steps conducted as part of the sequential extraction process. Because 100 Area soils are known to contain mica minerals, any treatment scheme (attrition scrubbing or chemical extraction) should include chemical components to mobilize as much of the cesium that is specifically-adsorbed cesium from wedge sites of micaceous minerals that are embedded in the bulk rock matrices.

Europium appears to be yet another radionuclide contaminant typically detected in 100 Area soils. This element belongs to the lanthanide group consisting of 14 elements with atomic numbers ranging from 58 (Cerium) to 71 (Lutetium). These lanthanide elements exist principally as trivalent cations and therefore form very strong complexes with many inorganic and organic ligands. Therefore, complexing agents can readily mobilize europium from the surfaces of contaminated soils. The attrition scrubbing (with electrolyte) and chemical extraction data on 2 to 0.25-mm fractions from 116-D-1B (Batch III) soil indicated that substantial fractions (>90%) of europium were removed from this soil. One of the factors in achieving these removal efficiencies appeared to be the use of complexing agents both in scrubbing electrolyte and in chemical extracts. The data obtained in these tests indicated that substantial fractions of europium can be removed if this element exists mainly as a surface contaminant (adsorbed on aluminosilicate and iron oxide coatings on soil particle surfaces)

The soils from the 100 Area that were analyzed in this study also contain  $^{137}\text{Co}$  as one of the contaminants. Cobalt is a first transition group element that includes among others

chromium, manganese, and iron. Therefore cobalt exhibits geochemical properties that are typical of these elements. Cobalt exists principally in a divalent state and forms moderately strong complexes with many organic and inorganic ligands. In soils, cobalt exists mainly in coprecipitated and adsorbed forms with oxides and hydroxides of iron and manganese. The results of sequential extraction experiments indicated that substantial amounts of cobalt was mobilized in extractive steps that apparently dissolved oxides and hydroxide precipitates of aluminum, iron, and manganese. Both attrition scrubbing and chemical extraction treatments (consisting of organic complexing agents) of 116-D-1B (Batch III) soil indicated that significant quantities (>80%) of radioactive cobalt were mobilized from this soil.

## 11.2 SOIL WASHING PARAMETERS

It is well established that soil washability depends on a number of physical, chemical and mineralogical properties of contaminated soils. These factors among many include the type, concentration, and the distribution of contaminant, TOC, pH, CEC, extractable oxide and hydroxide content, particle-size distribution and clay content. Even though soil washing has been used as a proven remediation technology for a number of years and the factors that affect washability are well recognized, no quantifiable relationships have been developed. The principal reason for this deficiency appears to be the lack of sufficient data to correlate properties of the contaminants and soils. If such data were collected, efforts could be directed to formulate first approximation predictive models for soil washing. Such models may involve multiple regressive relationship between removability of a contaminant and a number of physical, chemical and mineralogical parameters of soils. Based on these multiple regression models, a few key parameters that have significant functional relationships could be identified and used in a more refined predictive model.

Based on our soil washing bench-scale tests involving three soil samples from 100 Area, we have started the process of identifying several important parameters that affect the removal of three contaminant radionuclides. Some of these parameters include the particle-size distribution, radionuclide activity and their distribution among soil fractions, the presence of aluminosilicate and iron oxide coatings on sand and gravel particles and the quantity of micaceous minerals in the soil matrix. Because our data base involves soils from only two locations, more data are needed before other key factors that may affect soil washing are identified and quantified. A conceptual soil washing model developed for 100 Area soils will be a useful tool to predict the washability of soils in this area based on a few selected parameters. A reliable model can provide a cost and time effective path for soil remediation.

## 11.3 RESULTS OF THE FEASIBILITY TESTS

The sample of 116-C-1, Batch I material contained very low activities of all three radionuclides ( $^{137}\text{Cs}$ ,  $^{152}\text{Eu}$ , and  $^{60}\text{Co}$ ) in all the size fractions coarser than the silt and clay fractions. The silt and clay fraction constituting only 3% of the soil mass contained  $^{152}\text{Eu}$ ,

and  $^{60}\text{Co}$  activities of 50 and 18 pCi/g respectively. Because this soil contained, in the bulk, levels of radionuclides that were well below the TPL, we did not conduct any additional soil-washing tests on this material. This means that soils from other parts of 116-C-1 trench that are similar to Batch I material do not need to be washed.

The radionuclide data for soil samples from the 116-C-1 trench (Batch II) indicated that the gravel-fractions of samples obtained from the vicinity of the inlet had significantly higher activities of  $^{60}\text{Co}$ ,  $^{137}\text{Cs}$ , and  $^{152}\text{Eu}$  than the sample collected from the middle of the trench (Batch I) and also exceeded the proposed TPL. Therefore, a number of soil-washing tests were conducted on the gravel fraction of the Batch II soil that included autogenous grinding with electrolyte or chemical extractant, and static leaching with extractant. The data from these tests showed that in all cases substantial amounts of  $^{152}\text{Eu}$ , and  $^{60}\text{Co}$  were removed resulting in residual activities that were below TPL for these radionuclides. Autogenous grinding of the Batch II gravels with a chemical extractant (Extractant II) achieved the best removal rate for  $^{137}\text{Cs}$  (57%). However, the residual Cs activity in the treated gravels still remained well above the TPL for this contaminant. The inherent high activity in the Batch II material requires that at least >93 % reduction in Cs activity must be achieved for the soil to meet the TPL. Other potential tests that may achieve the required level of reduction in Cs activity may include leaching with hot mineral acids such as nitric and hydrochloric acid, and autogenous grinding in hot chemical extractant. It is important to note that even though the autogenous grinding experiments on 116-C-1 (Batch II) gravels did not result in cesium activity reductions below the TPL, such grinding may prove effective on gravels from other trenches with lower levels of cesium contamination.

The bulk soil material from the 116-D-1B (Batch III) trench contained activity levels for two radionuclides,  $^{137}\text{Cs}$  (103 pCi/g) and  $^{152}\text{Eu}$  (84 pCi/g) that exceeded proposed TPL (See Table 5-1). The radionuclide distribution data for this soil show that the levels of activity increased with decreasing particle-size. The wet-sieving data indicated that volume reductions ranging from 80 -90 % can be achieved for this soil if the activities of  $^{137}\text{Cs}$  and  $^{152}\text{Eu}$  in the sand fraction (2 to 0.25-mm size fraction) were reduced at least by 50% and 40%, respectively. Therefore, additional soil-washing tests such as attrition scrubbing and chemical extraction were conducted on this specific size fraction. The results indicated that the activity reduction goals can be met by using either of two different treatments. A two-stage attrition scrubbing conducted in an electrolyte medium, and a single chemical extraction with a proprietary extractant effectively reduced the activities of both  $^{137}\text{Cs}$  and  $^{152}\text{Eu}$  to the required levels.

Each of these effective treatment processes has a number of advantages and disadvantages. The advantages of two-stage attrition scrubbing with electrolyte include a grinding process that is conducted at ambient temperature, and the scrubbing that is conducted at high pulp densities (about 83%) thus requiring only a small amount of electrolyte. Third, the processing time is relatively short (15 to 30 min). Finally, attrition scrubbing is a relatively efficient unit operation. The disadvantages of this process include (i) the generation of contaminated fines (10 to 13 % by wt) that need to be disposed of, (ii)

the need to remove fines between each scrubbing stage to prevent reduction in scrubbing efficiency, and (iii) process wash water with high content of suspended solids (turbidities of about 8000 NTU) and residual electrolyte that need treatment before recycling.

The principal advantage of the chemical extraction process is that the process can be optimized to remove cesium to the level required to just meet the TPL therefore, the chemical costs can be reduced for specific circumstances. Second, the extractants contain fairly innocuous chemicals, and also remove substantial fractions of europium and cobalt. Third, the chemical extraction is a single step treatment process that does not require any intermediate processing step. The principal disadvantages are that the extractive process is conducted above ambient temperatures (80 to 96° C), the spent extractant contains high levels of soluble cesium activities that probably require precipitation and/or ion exchange treatment, and chemical extraction processes are relatively costlier than physical treatment processes such as size-fractionation and attrition scrubbing.

The soils tested in this study represent a textural range from poorly-graded gravel (Batch II) to well-graded sand with silt (Batch III). These soils also contain radionuclide activities that ranged from relatively low (Batch I) to moderately high (Batch II). Therefore, the radionuclide removal efficiencies of various soil-washing treatments also showed significant differences. These data agree with published data that a number of factors such as the physical, chemical and mineralogical properties, and the type and level of radionuclide contamination, control the choice and effectiveness of the various soil-treatment schemes. The literature data and the results of this study indicate that by choosing appropriate soil-washing schemes it is possible to remediate different types of radionuclide contaminated soils. As a final step however, these successful soil-washing schemes need to be analyzed for their cost effectiveness as compared to other remediation technologies.

## 11.4 SOIL-WASHING PROCESS OPTIONS FOR 116-D-1B SOIL

### 11.4.1 Physical Soil-Washing Process Options

At least three physical soil-washing options can be evaluated based on the data collected on particle size distribution levels of contamination, and the results of attrition scrubbing on sand fractions (2 to 0.25-mm size fraction) of 116-D-1B (Batch III) soil. The results of the tests and the processing options are listed in Table 11-1 and 11-2. These tables show the mass and contaminant distribution in four size fractions namely, >2-mm (+10 mesh), 2 to 0.25-mm (-10 +60 mesh), 0.25 to 0.074-mm (-60 +200 mesh), and <0.074-mm (-200 mesh). Calculated average radionuclide activities in cumulative coarser and finer size fractions are also tabulated.

If no processing is considered for the soil in the 116-D-1B trench, all the excavated material containing average activities of 103 pCi/g of <sup>137</sup>Cs, 84 pCi/g of <sup>152</sup>Eu, and 6 pCi/g of <sup>60</sup>Co needs to be disposed. The first soil washing process option to be considered was wet-screening (Table 11-1). Computations showed that the quantities of screened material

Table 11-1. Wet-Sieving Processing Option for 116-D-1B (Batch III) Soil.

## Cs-137

Fraction	Size mm	Size Distribution			Average Contamination Levels			No Processing		Simple Wet Screening					
		Wt. in Fraction	Cum. in Fraction & Finer	Cum Wt. Larger	In Fraction	In Fraction & Finer	In Coarser Fractions			10M Cut (2mm)		80M Cut (0.25mm)		200M Cut (0.074mm)	
		Wt. %	pCi/g	Wt. %	pCi/g	Wt. %	pCi/g	Wt. %	pCi/g	Wt. %	pCi/g	Wt. %	pCi/g		
-200M	0.074	7.1	7.1	92.9	590	590.0	88.3	To ERDF 100	103	To ERDF 53.1	185.2	To ERDF	499	To ERDF	590
-80+200M	0.25	3.7	10.8	89.2	325	499.2	55.6					Backfill	66		
-10+80M	2	42.3	53.1	46.9	105	185.2	11.0					Backfill	56		
+10M	10	46.9	100	0.0	11	103.5						Backfill	11		
Total		100													

## Eu-152

Fraction	Size mm	Size Distribution			Average Contamination Levels			No Processing		Simple Wet Screening					
		Wt. in Fraction	Cum. in Fraction & Finer	Cum Wt. Larger	In Fraction	In Fraction & Finer	In Coarser Fractions			10M Cut (2mm)		80M Cut (0.25mm)		200M Cut (0.074mm)	
								Wt. %	pCi/g	Wt. %	pCi/g	Wt. %	pCi/g	Wt. %	pCi/g
-200M	0.074	7.1	7.1	92.9	819	819.0	27.5	To ERDF 100	84	To ERDF 53.1	155.9	To ERDF 10.8	499	To ERDF 7.1	590
80 + 200M	0.25	3.7	10.8	89.2	117	578.5	23.8							Backfill 92.9	28
-10 + 60M	2	42.3	53.1	46.9	48	155.9	2.0							Backfill 89.2	24
+10M	10	46.9	100	0.0	2	83.7						Backfill 46.9	2		
Total		100													

## Co-60

Fraction	Size mm	Size Distribution			Average Contamination Levels			No Processing		Simple Wet Screening						
		Wt. in Fraction	Cum. in Fraction & Finer	Cum Wt. Larger	In Fraction	In Fraction & Finer	In Coarser Fractions			10M Cut (2mm)		80M Cut (0.25mm)		200M Cut (0.074mm)		
								Wt. %	pCi/g	Wt. %	pCi/g	Wt. %	pCi/g	Wt. %	pCi/g	
-200M	0.074	7.1	7.1	92.9	49	49.0	2.3	To ERDF 100	6	To ERDF 53.1	9.6	To ERDF 10.8	499	To ERDF	7.1	590
60 + 200M	0.25	3.7	10.8	89.2	10	36.6	1.9							Backfill	92.9	2
-10 + 80M	2	42.3	53.1	46.9	3	9.6	1.0							Backfill	89.2	2
+10M	10	46.9	100	0.0	1	5.6						Backfill	46.9	1		
Total		100														

Table 11-2. Wet-Sieving and Attrition Scrubbing Options for 116-D-1B (Batch III) Soil.

## Cs-137

Fraction	Size mm	Size Distribution			Average Contamination Levels			Attrition Scrubbing w/ Water			Attrition Scrubbing w/ Electrolyte		
		Wt. in Fraction	Cum. in Fraction & Finer	Cum. Wt. Larger	In Fraction	In Fraction & Finer	In Coarser Fractions	Contamination Red. = 33% Fines Generation = 12%	Wt. %	pCi/g	Contamination Red. = 48% Fines Generation = 12%	Wt. %	pCi/g
-200M	0.074	7.1	7.1	92.9	590	590.0	88.3	Soil Washing Feed (+80-10M)	42.3	105	Soil Washing Feed (+80-10M)	42.3	105
								SW Fines	5.6	337	SW Fines	5.1	475
								Cleaned Soil (+60M)	36.8	70	Cleaned Soil (+60M)	37.2	55
-60+200M	0.25	3.7	10.8	88.2	325	499.2	55.6	To ERDF			To ERDF		
								-60M (-0.25mm)	10.8	499	-60M (-0.25mm)	10.8	499
								SW Fines	5.6	337	SW Fines	5.1	475
								Total	16.3	444	Total	15.9	491
-10+60M	2	42.3	53.1	46.9	105	185.2	11.0	Beck III			Beck III		
								+10M (+2mm)	46.9	11	+10M (+2mm)	46.9	11
								Cleaned Soil (+60M)	36.8	70	Cleaned Soil (+60M)	37.2	55
								Total	83.7	37.1	Total	84.1	30.3
+10M	10	46.9	100	0.0	11	103.5							
Total		100											
										TPL = 30			

## Eu-152

Fraction	Size mm	Size Distribution			Average Contamination Levels			Attrition Scrubbing w/ Water			Attrition Scrubbing w/ Electrolyte		
		Wt. in Fraction	Cum. in Fraction & Finer	Cum. Wt. Larger	In Fraction	In Fraction & Finer	In Coarser Fractions	Contamination Red. = 71% Fines Generation = 13%	Wt. %	pCi/g	Contamination Red. = 90% Fines Generation = 12%	Wt. %	pCi/g
-200M	0.074	7.1	7.1	92.9	619	619.0	27.5	Soil Washing Feed (+80-10M)	42.3	48	Soil Washing Feed (+80-10M)	42.3	48
								SW Fines	5.5	276	SW Fines	5.1	365
								Cleaned Soil (+60M)	36.8	14	Cleaned Soil (+60M)	37.2	5
-60+200M	0.25	3.7	10.8	88.2	117	578.5	23.8	To ERDF			To ERDF		
								-60M (-0.25mm)	10.8	499	-60M (-0.25mm)	10.8	499
								SW Fines	5.5	276	SW Fines	5.1	365
								Total	16.3	424	Total	15.9	458
-10+60M	2	42.3	53.1	46.9	48	155.9	2.0	Beck III			Beck III		
								+10M (+2mm)	46.9	2	+10M (+2mm)	46.9	2
								Cleaned Soil (+60M)	36.8	14	Cleaned Soil (+60M)	37.2	5
								Total	83.7	7.2	Total	84.1	3.2
+10M	10	46.9	100	0.0	2	83.7							
Total		100											
										TPL = 15			

## Co-60

Fraction	Size mm	Size Distribution			Average Contamination Levels			Attrition Scrubbing w/ Water			Attrition Scrubbing w/ Electrolyte		
		Wt. in Fraction	Cum. in Fraction & Finer	Cum. Wt. Larger	In Fraction	In Fraction & Finer	In Coarser Fractions	Contamination Red. = 80% Fines Generation = 13%	Wt. %	pCi/g	Contamination Red. = 80% Fines Generation = 12%	Wt. %	pCi/g
-200M	0.074	7.1	7.1	92.9	48	49.0	2.3	Soil Washing Feed (+80-10M)	42.3	3	Soil Washing Feed (+80-10M)	42.3	3
								SW Fines	5.5	19	SW Fines	5.1	21
								Cleaned Soil (+60M)	36.8	1	Cleaned Soil (+60M)	37.2	1
-60+200M	0.25	3.7	10.8	88.2	10	36.8	1.9	To ERDF			To ERDF		
								-60M (-0.25mm)	10.8	499	-60M (-0.25mm)	10.8	499
								SW Fines	5.5	19	SW Fines	5.1	21
								Total	16.3	337	Total	15.9	348
-10+60M	2	42.3	53.1	46.9	3	9.6	1.0	Beck III			Beck III		
								+10M (+2mm)	46.9	1	+10M (+2mm)	46.9	1
								Cleaned Soil (+60M)	36.8	1	Cleaned Soil (+60M)	37.2	1
								Total	83.7	0.8	Total	84.1	0.8
+10M	10	46.9	100	0.0	1	5.8							
Total		100											
										TPL = 7			

that can be returned as backfill and their average radionuclide contents depend on the choice of screen used for the sieving operation. For instance, using the coarsest screen (10 mesh), approximately 47% of the soil (>2-mm size fraction) can be recovered as backfill material. It is important to note that the activities of all three radionuclides in the backfill fraction are below TPLs. If a finer mesh (60 mesh) is used as the cut off point between backfilling and disposal, about 89% of the mass of soil can be recovered as the backfill material. However, the average activities of both  $^{137}\text{Cs}$  (56 pCi/g), and  $^{152}\text{Eu}$  (24 pCi/g) in the backfill fraction would exceed the TPLs for both these radionuclides. Using 200 mesh as the cut off point, bulk of the soil (about 93%) can be recovered with still higher activities of  $^{137}\text{Cs}$  (66 pCi/g), and  $^{152}\text{Eu}$  (28 pCi/g) that exceed the TPLs. Therefore, using only wet-sieving, no more than 47% of the bulk soil can be recovered as clean backfill.

The second processing option consists of wet-sieving followed by attrition scrubbing the 2 to 0.25-mm size fraction in two separate stages with water. After screening out the fines (-60 mesh material), the scrubbed sand fraction can be composited with +10 mesh material for backfilling. Mass and activity calculations based on this option showed that about 84% of the bulk soil can be recovered with average activities  $^{152}\text{Eu}$  (7.2 pCi/g), and  $^{60}\text{Co}$  (about 0.8 pCi/g) that are well below the TPLs. Only the average activity of  $^{137}\text{Cs}$  (37 pCi/g) in this composite exceeded the TPL of 30 pCi/g.

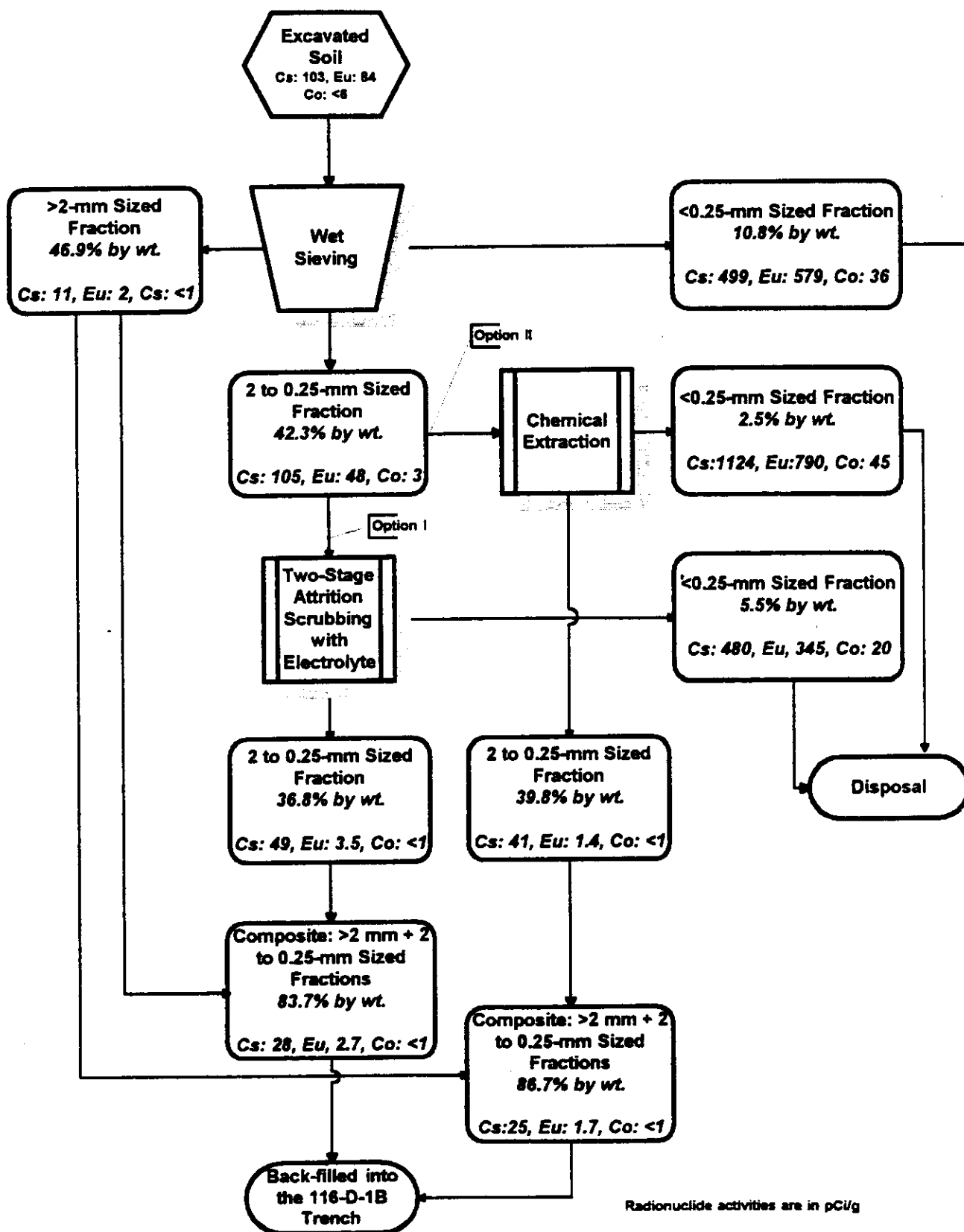
The third processing option is similar to the second option except that an electrolyte is used as the scrubbing medium. Same quantities (about 84%) of backfill material can be recovered using either second or third processing options. The main difference however, will be the lower residual radionuclide activities of backfill material resulting from the third processing option. The calculated average activities in the third option backfill material were 30.3, 3.2, and about 0.8 pCi/g for  $^{137}\text{Cs}$ ,  $^{152}\text{Eu}$ , and  $^{60}\text{Co}$  respectively. The calculated average activity of  $^{137}\text{Cs}$  in this backfill material is about equal to the TPL of 30 pCi/g, and the activities of  $^{151}\text{Eu}$ , and  $^{60}\text{Co}$  are well below the TPLs if 15 and 7 pCi/g respectively.

Therefore, among the three physical soil-washing process options for the 116-D-1B soil, the best option that results in a major mass of clean backfill material is the one that includes two-stage scrubbing of sand fraction with an electrolyte. We should also note that the bench-scale test data for the 116-D-1B (Batch III) soil indicated that using a chemical extraction step as part of an overall soil-washing scheme also results major mass of clean backfill material (about 87% by wt).

#### 11.4.2 A Preliminary Soil-Washing Scheme for 116-D-1B Soil

Based on data from the laboratory-scale tests reported here, a general scheme was devised for soil-washing the 116-D-1B soil (Fig 11-1). As the initial step, the scheme involves size-fractionating the soil into three fractions namely +10 mesh (>2-mm), -10 to +60 mesh (2 to 0.25-mm) and -60 mesh (<0.25-mm). The finest fraction (about 10.8% by mass) containing higher radionuclide activities is disposed. The 2 to 0.25-mm fraction can be treated either by using attrition scrubbing in two-stages with an electrolyte or batch extracting with the optimized proprietary aqueous extractant II. The higher activity fines

Figure 11-1. A Soil-Washing Scheme for 116-D-1B Trench Soils.





generated from either of these treatments (about 2.5 to 5.5 % of the whole soil mass) are diverted for disposal. The treated 2 to 0.25-mm fraction (37- 40% of the soil mass) when composited with the >2-mm fraction results in total recovery of approximately 85 % of originally excavated soil mass that can be backfilled into the trench. This proposed soil-washing scheme was used to formulate process flow sheets and to evaluate the cost effectiveness of this proposed soil-washing scheme.

### 11.5 PROPOSED PILOT-SCALE SOIL-WASHING TESTS ON 116-D-1B SOIL

The bench-scale soil-washing treatability test data on 116-D-1B soil was used by EEC (Enserch Environmental Corporation) to develop a process flow sheet for conducting pilot-scale soil-washing tests. The process flow scheme (Figure 11-2) is based on the third physical processing option (see section 11.4.1) that incorporates wet-screening and two-stage scrubbing with an electrolyte to achieve a clean backfill soil fraction. This flow scheme proposes using a soil-washing process that consists of various particle size fractionation unit operations. In this flow scheme, soil particle separations are conducted sequentially by using a grizzly, a primary screen, a trommel and a spiral classifier. The cleaned oversize fractions resulting from the first three screening operations diverted as part of the overall backfill material. The sand fraction from the spiral classifier is fed into a four-cell attrition scrubber. The sand fractions in this unit are scrubbed with an electrolyte for an appropriate residence time. Following this operation, the fines (-60 mesh) are screened out of the scrubbed material and the sand fraction is conveyed into a second attrition scrubbing unit and again scrubbed with an electrolyte for the required length of time. The clean material issuing from this second-stage scrubbing is washed and dewatered before being composited with the rest of the backfill material. The contaminated fines (-60 mesh) from various treatment stages are removed for disposal. The process flow scheme also includes other essential unit operations such as treatment of wash water and electrolyte, and recycling of treated aqueous waste streams.

The proposed pilot-scale tests to collect detailed data under field conditions are being planned for the summer of 1994.

### 11.6 PRELIMINARY COST ESTIMATES FOR SOIL WASHING OPTIONS

The cost of soil-washing depends on the processing options that are selected. Preliminary cost estimates were made by WHC using a number of assumptions regarding the cost of labor, analyses, maintenance, utilities and chemicals. These estimates provide a basis for relative cost comparison between various treatment options and direct disposal. These cost comparison data sheets are included (Appendix A).

## 11.7 SUPPLEMENTARY DATA REQUIREMENTS

The bench-scale feasibility tests indicated that there are a number of factors that affect the release of contaminants during the soil-washing processes. The qualitative factors that were recognized to be important included clay and iron oxide coatings, presence of micaceous minerals, and the depth of radionuclide contamination into the bulk interior of gravel-size material. However, these factors were not quantified and also their functional relationship if any, with the extent of radionuclide association and removal was not attempted. Because these factors are known to affect the removability of radionuclides from soils, it is useful to examine these factors and their relationship and to establish a predictive model for washability of the 100 Area soils.

The soil-washing feasibility data collected during this study provided a basis for designing a pilot-scale soil-washing test. However, a number of important questions need to be addressed further before the pilot-scale tests are conducted. One of these issues needing attention is the potential mobilization of regulated trace metals and radionuclides from the cleaned back-filled material. The washing treatments especially the chemical processes may perturb the normally tightly bound trace constituents and make them more susceptible for long-term release. Therefore extraction tests using the TCLP should be conducted on the cleaned backfill material. Another factor that should be examined, is the recyclability of aqueous waste streams such as wash water from particle classification operations and the electrolyte containing solutions from attrition scrubbing processes. It is necessary therefore to establish the number of times the treated liquid waste streams can be recycled before the build up of contaminants and macroions in reused solutions prevents further recycling. These studies should include a bench-scale study of waste water and electrolyte recycling, the rate of increase in concentrations of all major cations and anions, and the contaminant radionuclides ( $^{137}\text{Cs}$ ,  $^{152}\text{Eu}$ , and  $^{60}\text{Co}$ ) during the recycling process. This study should also include treatability tests on bleed-off waste streams for removal of suspended solids and soluble radionuclides through flocculation, precipitation and ion exchange. These tests are necessary because the cost effectiveness of soil-washing operations also depends on the recyclability of liquid waste streams.

Finally, the variability of contaminant activities and the bulk sediment particle-size distribution within the 116-D-1B trench should be further investigated prior to the pilot-scale tests to allow a more informed process flow sheet to be prepared. Our current data is based on only three soil samples (Two samples from 116-C-1 trench, and only one from 116-D-1B trench) shows a large range in particle size distribution [poorly-graded gravel (116-C-1, Batch II), to well-graded sand with silt (116-D-1B, Batch III), and a large range in  $^{137}\text{Cs}$  activities (726 pCi/g in Batch II, and 2 pCi/g in Batch I). It is likely that the 11-D-1B trench contains regions that have all these types of particle-size/contaminant pairs. Some knowledge of the locations and the extent of these various combinations will be useful in designing and conducting the pilot-scale tests.

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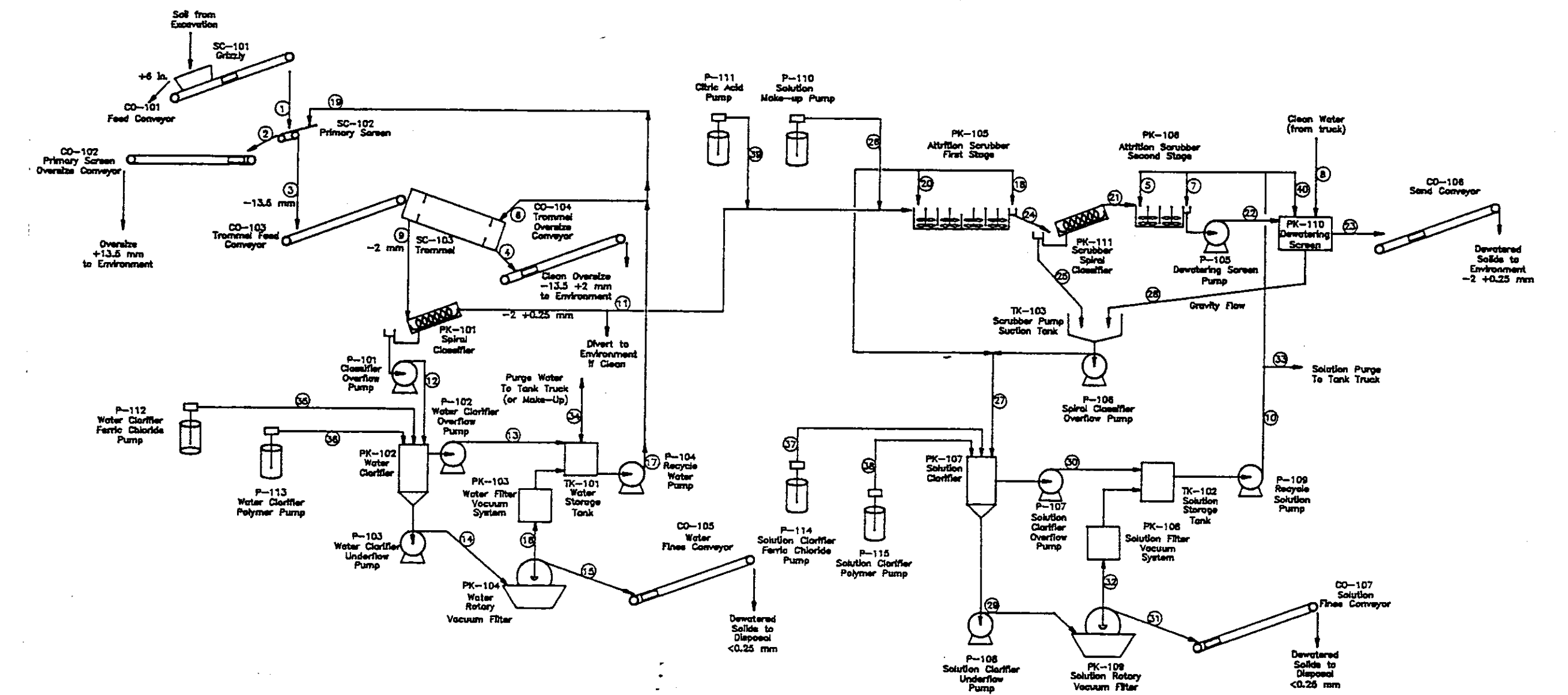


Figure 11-2. Process Flow Diagram for Pilot-Scale Tests on 116-D-1B (Batch III) Trench Soil

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ANY REVISION TO THIS DRAWING  
MUST BE MADE BY COMPUTER  
AND CHECKED

WESTINGHOUSE HANFORD COMPANY  
RICHLAND, WASHINGTON  
TASK# E-94-04

100 AREA SOIL WASHING TREATABILITY TEST  
PROCESS FLOW DIAGRAM

ENSERCH ENVIRONMENTAL CORPORATION  
Formerly Environmental Division of  
Ebasco Services, Inc.

DATE: 1/18/94

WHC-3403  
SK-04-04-101 REV. 1

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## 12.0 REFERENCES

- ASTM C 127-88, 1993, "Standard Test Method for Specific Gravity and Absorption of Coarse Aggregate," *Annual Book of ASTM Standards*, Vol. 4.02, American Society of Testing Materials, Philadelphia, Pennsylvania.
- ASTM C 702-87, 1993, "Standard Practice for Reducing Samples of Aggregate to Testing Size," *Annual Book of ASTM Standards*, Vol. 4.02, American Society of Testing Materials, Philadelphia, Pennsylvania.
- ASTM D 421-85, 1993, "Practice for Dry Preparation of Soil Samples for Particle-Size Analysis and Determination of Soil Constants," *Annual Book of ASTM Standards*, Vol. 4.08, American Society of Testing Materials, Philadelphia, Pennsylvania.
- ASTM D 422-63, 1993, "Standard Test Method for Particle-Size Analysis of Soils," *Annual Book of ASTM Standards*, Vol 4.08, American Society of Testing Materials, Philadelphia, Pennsylvania.
- ASTM D 854-83, 1993, "Standard Test Method for Specific Gravity of Soils," *Annual Book of ASTM Standards*, Vol. 4.08, American Society of Testing Materials, Philadelphia, Pennsylvania.
- ASTM D 2487-90, 1993, "Standard Test Method for Classification of Soils for Engineering Purposes," *Annual Book of ASTM Standards*, Vol 4.08, American Society of Testing Materials, Philadelphia, Pennsylvania.
- ASTM D 4129-88, 1993, "Standard Test Method for Total and Organic Carbon in Water by High Temperature Oxidation and by Coulometric Detection," *Annual Book of ASTM Standards*, Vol. 11.01, American Society of Testing Materials, Philadelphia, Pennsylvania.
- Baker, V. R., ed., 1981, "Catastrophic Flooding, the Origin of the Channeled Scabland." In *Benchmark Papers in Geology*, Vol 55, Dowden, Hutchinson, and Ross, Inc., Stroudsburg, Pennsylvania.
- Belzile, N., P. Lecomte, and A. Tessier, 1989, "Testing Readsorption of Trace Elements During Partial Chemical Extractions of Bottom Sediments." *Environmental Science and Technology* 23:1015-1020.
- Benson, D. W., J. L. Nelson, and G. J. Alkire, 1963, *Chemical and Physical Properties of 100 Area Soils*, General Electric Company, Richland, Washington.

- 9443221.0098
- Bowen, H.J.M., 1979, *Environmental Chemistry of the Elements*, Academic Press, New York.
- Dorian, J. J., and V. R. Richards, 1978, *Radiological Characterization of the Retired 100 Areas*, UNI-946, United Nuclear Industries, Inc, Richland, Washington.
- Freeman, H. D., M. A. Gerber, S. V. Mattigod, and R. J. Serne, 1993, *100 Area Soil Washing Bench-scale Test Procedures*, PNL-8520, Pacific Northwest Laboratory, Richland, Washington.
- Gardner, W. H., 1986, "Water Content," Chapter 21 in *Methods of soil Analysis, part I, Physical and Mineralogical Methods*, p 493-544, American Society of Agronomy-Soil Science Society of America, Madison, Wisconsin.
- Gombert, D., 1992, *Soil Washing Evaluation by Sequential Extraction for Test Reactor Area Warm Waste Pond*, Westinghouse Idaho Nuclear Company, Inc., Idaho Falls, Idaho
- Guzek, S. J., and J. G. Field, 1993, *100 Area Soil Washing*, Field Note Book, EFL-1069, Westinghouse Hanford Company, Richland, Washington.
- Horton, J. H., and E. L. Albenesius, 1976, "Volume Reduction of Plutonium-Contaminated Soil." *Nuclear Technology* 30:86-88.
- Jackson, M. L., 1974, "Soil Chemical Analysis-Advanced Course", 2nd ed. Published by the Author, Madison, Wisconsin.
- Mullineaux, D. R., R. E. Wilcox, W. F. Ebaugh, R. Fryxell, and M. Rubin, 1977, "Age of the Last Major Scabland Flood of Eastern Washington, as Inferred from Associated Ash Beds of Mount St. Helens Set S," *Geological Society of America Abstracts with Programs*, 5(7):1105.
- Murray, J. R., 1993, "Soil Washing Treatability Testing of Warm Waste Pond Soils at INEL." In *Proceedings of Soil Decon '93*, Oak Ridge National Laboratory, Oak Ridge, Tennessee.
- Navratil, J. D., and R. L. Kochen, 1982, *Decontamination of Soil containing Plutonium and Americium*, RFP-3139, DOE-TIC-4500, Rockwell International, Rocky Flats Plant, Golden, Colorado.
- Phillips, C. R., W. S. Richardson, C. Cox, and M. C. Eagle, 1993, "A Pilot Plant for the Remediation of Radioactive Contaminated Soils using Particle-Size Separation Technology." In *Proceedings of Soil Decon '93*, Oak Ridge National Laboratory, Oak Ridge, Tennessee.

- PNL 7-40-48 (Rev. 1), 1990, "Procedures and Quality Control for Energy Dispersive X-Ray Fluorescence Spectroscopy Using the BFP Approach with the KEVEX 0810A System," *Analytical Chemistry Laboratory Compendium*, PNL-MA-599, Technical Procedure, Pacific Northwest Laboratory, Richland, Washington.
- PNL-ALO-105 (Rev. 0), 1992, "Procedure for Preparation of Samples to be Counted by Gamma-Ray Spectroscopy," *Analytical Chemistry Laboratory Compendium*, PNL-MA-599, Technical Procedure, Pacific Northwest Laboratory, Richland, Washington.
- PNL-ALO-106 (Rev. 0), 1990, "Acid Digestion for Preparation of Samples for Radiochemical Analysis," *Analytical Chemistry Laboratory Compendium*, PNL-MA-599, Technical Procedure, Pacific Northwest Laboratory, Richland, Washington.
- PNL-ALO-213 (Rev. 0), 1991, "Mercury in Water, Solids, and Sludges by Manual Cold Vapor Technique," *Analytical Chemistry Laboratory Compendium*, PNL-MA-599, Technical Procedure, Pacific Northwest Laboratory, Richland, Washington.
- PNL-ALO-280 (Rev. 0), 1990, "Inductively Coupled Plasma-Mass Spectrometric (ICP-MS) Analysis," *Analytical Chemistry Laboratory Compendium*, PNL-MA-599, Technical Procedure, Pacific Northwest Laboratory, Richland, Washington.
- PNL-ALO-282 (Rev. 0), 1991, "Determination of Uranium Concentration/Isotopic Composition Using ICP-MS," *Analytical Chemistry Laboratory Compendium*, PNL-MA-599, Technical Procedure, Pacific Northwest Laboratory, Richland, Washington.
- PNL-ALO-417 (Rev. 0), 1993, "Separation of Americium and Plutonium by Extraction Chromatography for Tank Samples," *Analytical Chemistry Laboratory Compendium*, PNL-MA-599, Technical Procedure, Pacific Northwest Laboratory, Richland, Washington.
- PNL-ALO-463 (Rev. 0), 1992, "Beta Counting Procedure," *Analytical Chemistry Laboratory Compendium*, PNL-MA-599, Technical Procedure, Pacific Northwest Laboratory, Richland, Washington.
- PNL-ALO-464 (Rev. 0), 1992, "Procedure for Gamma Counting and Data Reduction in the Low-Level Counting Room, 329 Building," *Analytical Chemistry Laboratory Compendium*, PNL-MA-599, Technical Procedure, Pacific Northwest Laboratory, Richland, Washington.
- PNL-ALO-465 (Rev. 0), 1992, "Strontium-90 Analysis (Oxalate-Nitric Acid Method)," *Analytical Chemistry Laboratory Compendium*, PNL-MA-599, Technical Procedure, Pacific Northwest Laboratory, Richland, Washington.

- PNL-ALO-466 (Rev. 0), 1992, "Procedure for Plutonium Separation and Initial Americium Separation by Anion Exchange," *Analytical Chemistry Laboratory Compendium*, PNL-MA-599, Technical Procedure, Pacific Northwest Laboratory, Richland, Washington.
- PNL-ALO-468 (Rev. 0), 1992, "Procedure for Electroplating Plutonium, Americium and Uranium onto Counting Disks," *Analytical Chemistry Laboratory Compendium*, PNL-MA-599, Technical Procedure, Pacific Northwest Laboratory, Richland, Washington.
- PNL-ALO-469 (Rev. 0), 1991, "Alpha Spectrometry," *Analytical Chemistry Laboratory Compendium*, PNL-MA-599, Technical Procedure, Pacific Northwest Laboratory, Richland, Washington.
- PNL-ALO-496 (Rev. 0), 1993, "Precipitation Plating of Actinides for High-Resolution Alpha Spectrometry," *Analytical Chemistry Laboratory Compendium*, PNL-MA-599, Technical Procedure, Pacific Northwest Laboratory, Richland, Washington.
- Richardson, W. S., T. B. Hudson, J. G. Wood, and C. R. Phillips, 1989, "Characterization and Washing Studies on Radionuclide Contaminated Soils." In *Superfund '89 Proceedings on the 10th Conference*, The Hazardous Materials Control Research Institute, Silver Spring, Maryland.
- Sawhney, B. L., 1964, "Sorption and Fixation of Microquantities of Cesium by Clay Minerals: Effect of Saturating Cations," *Soil Science Soc. Am Proc.* 28:183-186.
- Scott, A. D. and S. J. Smith, 1987, "Sources, Amounts, and Forms of Alkali Elements in Soils," in *Advances in Soil Science*, 6:101-147. Springer-Verlag, New York.
- Shigley, J. E., and C. R. Mischke, 1989, "Mechanical Engineering Design," McGraw Hill, New York.
- Soil Decon Task Group, 1993, "Removal of Uranium-contaminated Soils Phase I: Bench-Scale Testing," Oak Ridge National Laboratory, Oak Ridge, Tennessee.
- Stevens, J. R., and D. W. Rutherford, 1982, *Separation of Transuranic Radionuclides from Soil by Vibratory Grinding*, RFP-3296, DOE/TIC-4500, Rockwell International, Rocky Flats Plant, Golden, Colorado.
- Stevens, J. R., R. L. Kochen, and D. W. Rutherford, 1982, *Comparative Scrub Solution Tests for Decontamination of Transuranic Radionuclides from Soils*, RFP-3161, DOE/TIC-4500, Rockwell International, Rocky Flats Plant, Golden, Colorado.
- Thomas, G. W., 1986, "Exchangeable Cations," Chapter 9 in Methods of Soil Analysis, Part 2, Chemical and Microbiological Properties, p. 159-165, American Society of Agronomy-Soil Science Society of America, Madison, Wisconsin.



- Triay, I. R., G. K. Bayhurst, and A. J. Mitchell, 1993, *Report on the Effectiveness of Flocculation for The Removal of  $^{239}\text{Pu}$  from The RFP Pond Water*, LA-UR-93-1550, Los Alamos National Laboratory, Los Alamos, New Mexico.
- U.S. Department of Energy, Richland Operations Office (DOE-RL), 1988, *Final Status Post-Closure Permit Application: 183-H Solar Evaporation Basins*, DOE/RL-88-09, U.S. Department of Energy, Richland, Washington.
- U.S. Department of Energy, Richland Operations Office (DOE-RL), 1992a, *100 Area Feasibility Study Phases 1 and 2*, DOE/RL-92-11, Decisional Draft, U.S. Department of Energy, Richland, Washington.
- U.S. Department of Energy, Richland Operations Office (DOE-RL), 1992b, *100 Area Soil Washing Treatability Test Plan*, DOE/RL-92-51, Rev. 0, U.S. Department of Energy, Richland, Washington.
- U.S. Department of Energy, Richland Operations Office (DOE-RL), 1992c, *Hanford Site Soil Background*, DOE/RL-92-24, U.S. Department of Energy, Richland, Washington.
- U.S. Department of Energy, Richland Operations Office (DOE-RL), 1992d, *Treatability Study Program Plan*, DOE/RL-92-48, Decisional Draft, U.S. Department of Energy, Richland, Washington.
- U.S. Environmental Protection Agency (EPA), 1990, *Test Methods for Evaluating Solid Waste, Physical/Chemical Methods*, SW-846, 3rd Edition, U.S. Environmental Protection Agency, Washington, D.C.
- U.S. Department of Energy, Richland Operations Office (DOE/RL). 1993. 300-FF-1 Operable Unit Remedial Investigation Phase II Report: Physical Separation of Soils Treatability Study. DOE/RL-93-96, Draft, A, U.S. Department of Energy, Richland Operations Office, Richland, Washington.
- Westinghouse Hanford Company (WHC), 1988, *Environmental Compliance*, WHC-CM-7-5, Westinghouse Hanford Company, Richland, Washington.

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**APPENDIX A**

**COST ESTIMATES**

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## APPENDIX A

## COST ESTIMATES

**Approximate Order of Magnitude Soil Washing Cost Estimates (Thousands of \$ s)**  
**(For Comparison Only)**

Item	Wet Sieve	W/ Attrition	W/Chem Ext	Ref.
Purchase and Mobilize	2,075	3,385	3,275	Vendor Quotes times 2.5 for Hanford
X EUAC*	338	552	534	EUAC = (0.163)
Annual Cost				
Direct Labor	585	558	585	Assumed the same for all (DOE/RL 1993)**
Support Labor	385	358	358	
Materials	161	161	161	
Utilities	23	45	470	@ \$0.06/kWh
Analytical	171	171	171	Assumed the same for all (DOE/RL 1993)
Maintenance	121	121	121	
Ion Exchange Resin	190	190	190	Vendor quote, assumes 15 ft <sup>3</sup> /100 ton.
Chemicals	0	1,510	5,500	Assumes 242 lb/hr Electrolyte \$0.50/lb, Chem Ext \$1.82/lb.
ANNUAL COST	1,609	3,141	7,556	
TOTAL	1,947	3,693	8,090	ANNUAL + PURCHASE X EUAC
\$/TON	15.60	29.50	64.70	
* EUAC = Equivalent Uniform Annualized Cost ** DOE/RL, 1993				

Costs based on equipment designed to process 125,000 ton of soil/yr at 100 ton/hr.

**Case Study ROM Cost Estimates for Remediation of 125,000 ton/yr  
(For Comparison Only, Life Cycle Costs to be estimated later)**

**Remediation Costs Include:**

- Excavation Cost (assume \$125,000, \$1/ton)
- Soil Wash Cost (As shown in the Table)  
Based on 100 ton/hr, 5 d/w, 5 h/d, 50 w/y
- Haul and Disposal (May range to \$100/ton or more)

$$\text{Haul and Disposal} = \$ \text{ s/ton} \times 125,000 \times \% \text{ tons disposed}/100$$

**Eg. 1. Direct Disposal with \$100/ton haul and disposal cost**

**\$ Remediation =**

$$\begin{array}{l} \$1/\text{ton} \times 125,000 \text{ ton (excavation)} \\ \$0 \text{ (For Treatment)} \\ \$100 \times 125,000 \text{ ton} \times 100\%/100 \text{ (Haul and Dispose)} \\ \hline \$12,625,000 \\ \text{or } \$101/\text{ton} \end{array}$$

**Eg. 2. Remediation Cost for Wet Sieving with 47% cleaned.**

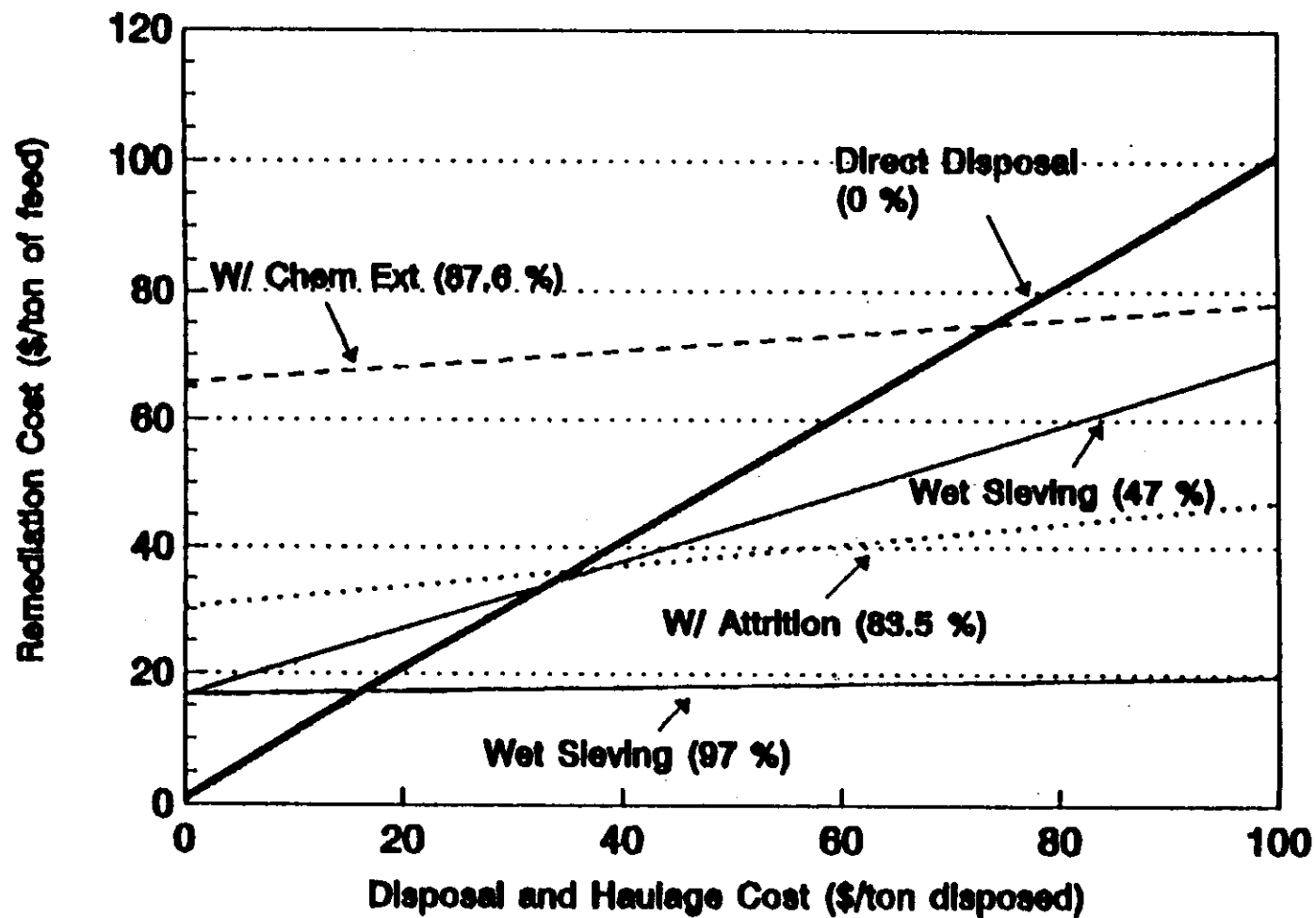
$$\begin{array}{l} \$1/\text{ton} \times 125,000 \text{ ton (excavation)} \\ \$15.60/\text{ton} \times 125,000 \text{ ton (wet sieving)} \\ \$100 \times 125,000 \text{ ton} \times 53\%/100 \text{ (Haul and Dispose)} \\ \hline \$8,700,000 \\ \text{or } \$69.6/\text{ton} \end{array}$$

**Calculations Assume:**

- Excavation cost is equivalent for all alternatives, \$1/ton.
- Analytical costs are the same for all alternatives
- Haul and disposal costs are the same for all alternatives

## Case Study: Direct Disposal vs. Treatment Cost Comparison

(Based on \$15.6/ton for Wet Slewing Only)



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